

## Chapter 3 Climate Change From Short-Lived Emissions Due to Human Activities

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This chapter addresses the four questions regarding short-lived species that were posed in the Prospectus for this Report:

**Question 1.** What are the impacts of the radiatively active short-lived species not explicitly the subject of prior CCSP assessments (SAP 2.1a: Scenarios of Greenhouse Gas Emissions and Atmospheric Concentrations)?

**Answer 1.** Uncertainties in emissions projections for short-lived species are very large, even for a particular storyline. For aerosols, these uncertainties are usually dominant, while for tropospheric ozone, uncertainties in physical processes are more important. Differences among modeled future atmospheric burdens and radiative forcing for aerosols are dominated by divergent assumptions about emissions from South and East Asia. Aerosol mixing, aerosol indirect effects, the influence of ecosystem-chemistry interactions on methane, and stratosphere-troposphere exchange all contribute to large uncertainties separate from the emissions projections.

**Question 2.** How do the impacts of short-lived species compare with those of the well-mixed greenhouse gases as a function of the time horizon examined?

**Answer 2.** By 2050, two of the three models show a global mean annual average enhancement of the warming due to long-lived greenhouse gases by 20-25% due to the radiatively active short-lived species (which are not being reported in SAP 2.1). One model shows virtually no effect from short-lived species. To a large extent, the inter-model differences are related to differences in emissions. Short-lived species may play a substantial role relative to well-mixed greenhouse gases out to 2100. One model finds that short-lived species can contribute 40% of the projected summertime warming in the central US.

**Question 3.** How do the regional impacts of short-lived species compare with those of long-lived gases in or near polluted areas?

**Answer 3.** The spatial distribution of radiative forcing is generally less important than the spatial distribution of climate sensitivity in predicting climate impact. Thus, both short-lived and long-lived species appear to cause enhanced climate responses in the same regions of high climate sensitivity rather than short-lived species having an enhanced effect primarily in or near polluted areas.

**Question 4.** What might be the climate impacts of mitigation actions taken to reduce the atmospheric levels of short-lived species to address air quality issues?

1705

1706 **Answer 4.** Regional air quality emission control strategies for short-lived pollutants have  
1707 the potential to substantially affect climate globally. Emissions reductions in the domestic  
1708 sector in developing Asia, and to a lesser extent in the surface transportation sector in  
1709 North America, appear to offer the greatest potential for substantial, simultaneous  
1710 improvement in local air quality and mitigation of global climate change.

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### 1712 **3.1 Introduction**

1713 In this chapter, we describe results from numerical simulations of 21<sup>st</sup> century climate,  
1714 with a major focus on the effects of short-lived gases and particulates. The calculations  
1715 incorporate results from three different types of models:

1716

- 1717 1. Integrated assessment models that produce emission scenarios for aerosols and for  
1718 ozone and aerosol precursor species.
- 1719 2. Global chemical composition models, which employ these emission scenarios to  
1720 generate concentrations for the short-lived radiatively active species.
- 1721 3. Global comprehensive climate models, which calculate the climate response to  
1722 the projected concentrations of both the short-lived and long-lived species. Box  
1723 1.1 outlines this sequence in detail.

1724

1725 The second part of Chapter 3, Section 3.2, is a discussion of the emission scenarios and  
1726 the models used to generate them, and the chemical composition models (sometimes  
1727 called chemical transport models) used to produce the global distributions of short-lived

species that help to drive the comprehensive climate models. Section 3.2 shows that, beginning with a single socio-economic scenario for the time evolution of long-lived (well-mixed) greenhouse gases, different assumptions about the evolution of the aerosols and precursor species lead to very different estimates of aerosol and ozone concentrations for the 21<sup>st</sup> century. We conclude that uncertainties in emissions projections for short-lived species are very large, even for a particular storyline. For aerosols, these uncertainties are usually dominant, while for tropospheric ozone, uncertainties in physical processes are more important.

The third part of Chapter 3, Section 3.3, discusses the three global comprehensive climate models (Geophysical Fluid Dynamics Laboratory (GFDL); Goddard Institute for Space Studies (GISS); National Center for Atmospheric Research (NCAR)) that have been used to calculate the impact of the short- and long-lived species<sup>34</sup> on the climate, focusing on the changes in surface temperature and precipitation. Supplementing the climate model results are calculations of the changes in radiative forcing<sup>35</sup> of the earth-atmosphere system. We find that by 2050, two of the three climate models show that radiatively active short-lived species enhance the global-mean annual-average warming due to long-lived greenhouse gases by 20-25%. One model shows virtually no effect from short-lived species. To a large extent, the inter-model differences are related to differences in emissions. One of the models has been extended to 2100. In that model,

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<sup>34</sup> We distinguish here between short-lived species (which have atmospheric lifetimes less than one month and are non-uniformly distributed) and long-lived species (which have lifetimes of a decade or more and are generally well mixed in the atmosphere).

<sup>35</sup> Radiative forcing is defined in Section 3.3.3; briefly, it measures the net change in the energy balance of the earth-atmosphere system with space associated with a change in composition of any radiatively active species present in the atmosphere

short-lived species play a substantial role, relative to the well-mixed greenhouse gases, in the surface temperature evolution out to 2100 and are responsible for 40% of the projected summertime warming in the central US.

The fourth part of Chapter 3, Section 3.4, discusses the effects of changes in regional aerosol and ozone and aerosol precursor emissions, using models that separate emissions by economic sector. The results show that regional air quality emission control strategies for short-lived pollutants have the potential to substantially affect climate at large-scales. Emission reductions from domestic sources in Asia, and to a lesser extent from surface transportation in North America, appear to offer the greatest potential for substantial, simultaneous improvement in local air quality and mitigation of global climate change.

## **3.2 Emission Scenarios and Composition Model Descriptions**

### **3.2.1 Emission Scenarios**

The long-lived (well-mixed) greenhouse gases included in this study were carbon dioxide (CO<sub>2</sub>), nitrous oxide (N<sub>2</sub>O), methane (CH<sub>4</sub>), and the minor species (chlorofluorocarbons, sulfur hexafluoride). Projected global mean values were prescribed following the A1B ‘marker’ scenario for all three modeling groups. Emissions for anthropogenic sources of aerosols and precursor species for all 3 composition model calculations were based on an international emission inventory maintained in the Netherlands (Olivier and Berdowski, 2001).

Though the three groups in this study all prescribed future emissions following a specific socio-economic scenario (A1B) that was highly studied in the latest report by the Intergovernmental Panel on Climate Change (IPCC), they used different emissions trends for the short-lived species. There are several reasons for the differences. For one, the A1B emissions projections only provide estimates of anthropogenic emissions, and each model used its own natural emissions (though these were largely held constant). Secondly, integrated assessment models, while using the same socio-economic storyline (A1B), provided a range of emission results (Nakicenovic *et al.*, 2000).

Two groups, GFDL and NCAR, used output from the AIM integrated assessment model (integrated assessment models are defined in Chapter 2, Section 2.1) while GISS used results from the IMAGE model. Though the emissions output generated from AIM was denoted the ‘marker’ scenario by the IPCC, it was noted that it did not represent the average, best, or median result, and that all integrated assessment model results should be treated equally. Finally, emissions for some species, such as carbonaceous aerosols, were not provided. This last issue motivated the GISS choice of the IMAGE model output, as it provided sufficient regional detail to allow carbonaceous aerosol emissions to be estimated consistently with the other species. Another complexity was the treatment of biomass burning emissions, which are partly natural and partly anthropogenic. In the GFDL model, biomass-burning emissions were assumed to be half-anthropogenic and half natural. The GISS model instead used biomass burning emissions projections from another inventory (Streets *et al.*, 2004).

1793 The result is a substantial divergence in the projected trends among the three models  
1794 (Figure 3.1, Table 3.1). For sulfur dioxide (SO<sub>2</sub>), the precursor to sulfate aerosol, the  
1795 emissions follow reasonably similar trajectories, with globally averaged increases until  
1796 2030 followed by decreases to 2050 and even further decreases to 2100. However, the  
1797 percentage increase is roughly double for GISS and NCAR as compared with GFDL.  
1798 Thus even two composition models using anthropogenic emission projections from the  
1799 same integrated assessment model show large differences in the evolution of their total  
1800 emissions, presumably owing to differences in the present-day emission inventories . At  
1801 2050, the GFDL model has substantially reduced emissions compared with 2000, while  
1802 the other models show enhanced emissions relative to 2000. A similar divergence in  
1803 projected sulfur-dioxide trends is present in the 2.1a stabilization emission scenarios  
1804 discussed in Chapter 2, with emissions decreasing dramatically (~70%) by 2050 in one  
1805 integrated assessment model (MINICAM) while decreasing only moderately (~20%) in  
1806 the two others, and even beginning to increase again after about 2040 in one of those two.  
1807  
1808 Differences are even more striking for carbonaceous aerosols emissions, which were not  
1809 provided by any of the integrated assessment models. We focus on black carbon (BC) as  
1810 the more important radiative perturbation. For this aerosol (and for organic carbon(OC)),  
1811 the GFDL composition model uses the IPCC recommendation to scale carbonaceous  
1812 aerosol emissions to carbon monoxide emissions, leading to substantial increases with  
1813 time (Figure 3.1, Table 3.1). However, many of the sources of carbon monoxide emission  
1814 are different from those of carbonaceous aerosols. The NCAR group did not simulate the  
1815 future composition of black and organic carbon based on emission projections, but

instead scaled their present-day distribution by the global factors derived for sulfur dioxide. The time evolution of black and organic carbon emissions in the NCAR model thus follows the same trajectory as that of SO<sub>2</sub>. On the other hand, the GISS group used emissions projections from (Streets *et al.*, 2004) based on energy and fuel usage trends from the IMAGE model (as for other species) and including expected changes in technology. This led to a substantial reduction in future emissions of carbonaceous aerosols.

For precursors of tropospheric ozone, there was again divergence among the models. The primary precursor in most regions, NO<sub>x</sub> (nitrogen oxides = NO + NO<sub>2</sub>), increased steadily in the projections used by GISS, while it peaked at 2030 and decreased slightly thereafter in the projections used at GFDL (Table 3.1). Hydrocarbons and carbon monoxide show analogous differences. Methane was prescribed according to the A1B “marker” scenario values for all three composition models. Thus ozone, in addition to the aerosols, was modeled in substantially different ways at the three centers.

The three models included projected changes in the same species, with the exception of nitrate, which only varied in the GISS model. As its contribution to total aerosol and total aerosol radiative forcing is small, at least in this GISS model, this particular difference was not significant in our results.

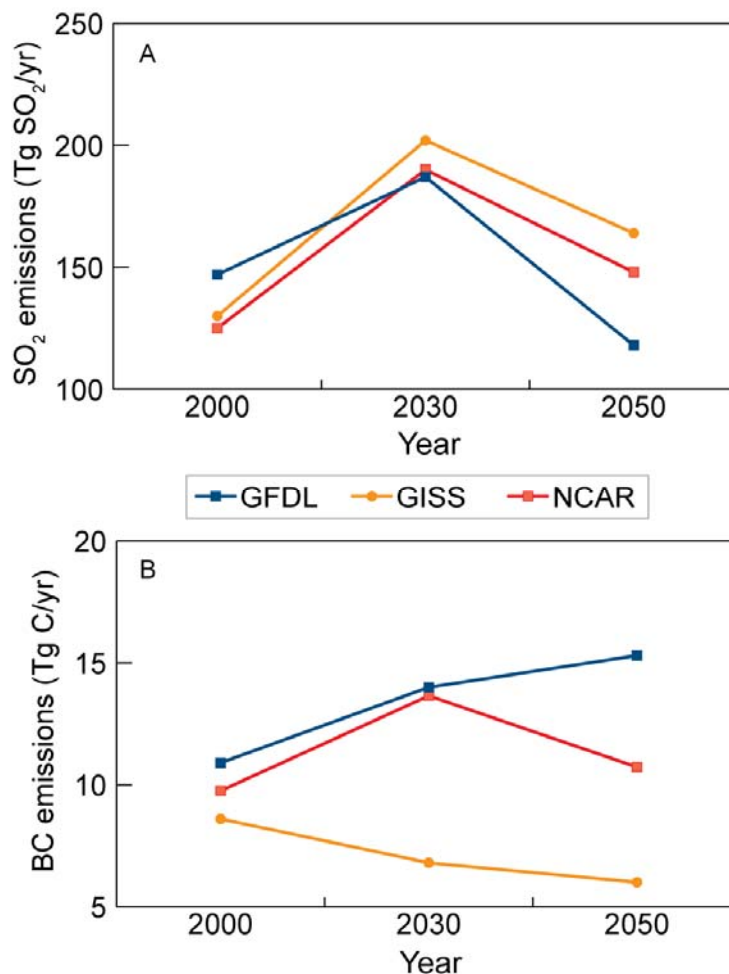


**Table 3.1. Global Emissions. Emissions include both natural and anthropogenic sources. Values in parentheses are changes relative to 2000.**

Species	Model	2000	2030	2050	2100
NO <sub>x</sub> (Tg N/yr)	GFDL	40	57 (43%)	54 (35%)	48 (20%)
	GISS	50.5	67.0 (33%)	77.5 (53%)	NA
BC (Tg C/yr)	GFDL	10.9	14.0 (28%)	15.3 (40%)	19.9 (83%)
	GISS	8.6	6.8 (-21%)	6.0 (-30%)	NA
OC (Tg C/yr)	GFDL	51.5	61.9 (20%)	66.5 (29%)	84.3 (%)
	GISS	69.5	57.0 (-18%)	58.3 (-16%)	NA
SO <sub>2</sub> (Tg SO <sub>2</sub> /yr)	GFDL	147	187 (27%)	118 (-20%)	56 (-62%)
	GISS	130	202 (55%)	164 (26%)	NA
	NCAR	125	190 (52%)	148 (18%)	NA
Dust (Tg/yr)	GFDL	2471	2471	2471	2471
	GISS	1580	1580	1580	NA

### 3.2.2 Composition Models

The chemical composition models used to produce short-lived species concentrations for the GFDL, GISS, and NCAR climate models were driven by the emissions projections discussed in Section 3.2.1. While the three models did not use identical present-day emissions, their anthropogenic emissions were based on the same international inventory (Olivier and Berdowski, 2001). The chemical composition simulations were run for one or two years, with the three-dimensional monthly mean concentrations and optical properties archived for use as off-line concentration fields to drive the climate model simulations discussed in Section 3.3. These simulations were all performed with present-day meteorology (values for temperature, moisture, and wind). Further details about the chemical composition models are provided in Appendix 1.



**Figure 3.1** A1B emissions trends used in the three models for SO<sub>2</sub> (top) and BC (bottom). Note that in the NCAR model, the present day black carbon distribution was scaled in the future rather than calculated from BC emissions. Scaling was chosen to mimic the global sulfur dioxide emissions, a 40% increase over 2000 at 2030, and 10% at 2050. The NCAR 2000 black carbon global emission is set at the average of the GISS and GFDL 2000 values, and follows this scaling in the future, for illustrative purposes.

### 3.2.2.1 Geophysical Fluid Dynamics Laboratory (GFDL)

Composition changes for the short-lived species in the GFDL experiments were calculated using the global chemical transport model MOZART-2 (Model for OZone And Related chemical Tracers, version 2.4), which has been described in detail previously (Horowitz *et al.*, 2003; Horowitz, 2006; and references therein). This model was used to generate the monthly average distributions of tropospheric ozone, sulfate, and black and organic carbon as a function of latitude, longitude, altitude, and time for

the emission scenarios discussed above. Simulated ozone concentrations agree well with present-day observations and recent trends (Horowitz, 2006). Overall, the predicted concentrations of aerosol are within a factor of two of the observed values and have a tendency to be overestimated (Ginoux *et al.*, 2006). Further details on the MOZART model are found in Appendix 3.1, in the section on Geophysical Fluid Dynamics Laboratory.

### 3.2.2.2 Goddard Institute for Space Studies (GISS)

The configuration of the GISS composition model used here has been described in detail in (Shindell *et al.*, 2007). In brief, the composition model PUCINI (Physical Understanding of Composition-Climate Interactions and Impacts) includes ozone and oxidant photochemistry in both the troposphere and stratosphere (Shindell *et al.*, 2006b), sulfate, carbonaceous and sea-salt aerosols (Koch *et al.*, 2006, 2007), nitrate aerosols (Bauer *et al.*, 2006), and mineral dust (Miller *et al.*, 2006a). Present-day composition results in the model are generally similar to those in the underlying chemistry and aerosol models. Further details on the PUCINI model resolution, composition, and performance are found in Appendix 3.1, in the section on Goddard Institute for Space Studies.

### 3.2.2.3 National Center for Atmospheric Research (NCAR)

For the climate simulations described in this section, present-day tropospheric ozone was taken from (Lamarque *et al.*, 2005a); beyond 2000, tropospheric ozone was calculated by T. Wigley using the MAGICC composition model (<http://www.cru.uea.ac.uk/~mikeh/software/magicc.htm>) forced by the time-varying

emissions of NO<sub>x</sub>, methane and volatile organic compounds (VOCs) and these average global values were used to scale the present-day distribution. Future carbonaceous aerosols are scaled from their present-day distribution (Collins *et al.*, 2001) by a globally uniform factor whose time evolution follows the global evolution of SO<sub>2</sub> emissions. Stratospheric ozone changes are prescribed following the study by (Kiehl *et al.*, 1999). Further details on the composition models used by NCAR are found in Appendix 3.1 in the section on National Center for Atmospheric Research.

### 3.2.3 Tropospheric Burden

The composition models each calculate time-varying three-dimensional distributions of the short-lived species (except for NCAR where 2030 and 2050 ozone, black carbon, and organic carbon were scaled based on their 2000 distributions). We compare these using the simple metric of the global mean annual average tropospheric burden (*i.e.* the total mass in the troposphere). As was the case with emissions, the differences between the outputs of the composition models are substantial (Table 3.2). The GFDL model has a 67% greater present-day burden of sulfate than the GISS model, for example. As the GFDL sulfur dioxide emissions were only 13% greater, this suggests that either sulfate stays in the air longer in the GFDL model than in the GISS model or sulfur dioxide is converted more efficiently to sulfate in the GFDL model.

1908 **Table 3.2 Global Burdens.** Values in parentheses are changes relative to 2000.

Species	Model	2000	2030	2050	2100
BC (Tg C)	GFDL	0.28	0.36 (29%)	0.39 (39%)	0.51
	GISS	0.26	0.19 (-27%)	0.15 (-42%)	NA
	NCAR		(40%)	(10%)	
OC* (Tg C)	GFDL	1.35	1.59 (18%)	1.70 (26%)	2.15
	GISS	1.65	1.33 (-19%)	1.27 (-23%)	NA
	NCAR		(40%)	(10%)	
Sulfate (Tg SO <sub>4</sub> <sup>-</sup> )	GFDL	2.52	3.21 (27%)	2.48 (-2%)	1.50 (-40%)
	GISS**	1.51	2.01 (33%)	1.76 (17%)	NA
	NCAR				
Dust (Tg)	GFDL	22.31	22.31	22.31	22.31
	GISS	34.84	34.84	34.84	NA
	NCAR				
Tropospheric Ozone (DU)	GFDL	34.0	38.4 (+13%)	39.3 (+16%)	38.2 (+12%)
	GISS	31.6	41.5 (31%)	47.8 (51%)	NA
	NCAR	28.0	41.5 (48%)	43.0 (54%)	NA

\*The organic carbon (OC) burdens include primary OC aerosols (with emissions as in above table) plus secondary OC aerosols (SOA). In the GFDL model, the global burden of SOA is 0.07 Tg C in this inventory. In the GISS model, organic carbon from SOA makes up ~24% of present-day OC emissions.

\*\*GISS sulfate burdens include sulfate on dust surfaces, which makes up as much as ½ the total burden.

1914 This can be tested by analyzing the atmospheric residence times of the respective models  
 1915 (Table 3.3). The residence time of sulfate is within ~10% in the two models, and in fact is  
 1916 slightly less in the GFDL model. This indicates that the conversion of sulfur dioxide  
 1917 (SO<sub>2</sub>) to sulfate must be much more efficient in the GFDL model for it to have a sulfate  
 1918 burden so much larger than the GISS model. This is clearly seen in the ratio between  
 1919 sulfate burden and SO<sub>2</sub> emissions (Table 3.4). This ratio can be analyzed in terms of the  
 1920 total sulfur dioxide burden (in Tg) per SO<sub>2</sub> emission (in Tg/yr); the change in SO<sub>2</sub> burden  
 1921 per SO<sub>2</sub> emission change, or alternatively in the percentage change in each. The latter is  
 1922 probably the most useful evaluation, as the fractional change will reduce differences  
 1923 between the starting points of the two models. We note that this metric is affected by both  
 1924 production and removal rates in the models. Table 3.4 shows clearly that the production  
 1925 of sulfate per Tg of sulfur emitted is much greater in the GFDL model than in the GISS

model, either because of differences in other sources of sulfate (*e.g.*, from dimethyl sulfate (DMS)) or difference in the chemical conversion efficiency of SO<sub>2</sub> to sulfate (versus physical removal of SO<sub>2</sub> by deposition).

**Table 3.3 Global mean annual average aerosol residence times (days)**

Species	Model	2000	2030	2050
BC	GFDL	9.4	9.4	9.3
	GISS	11.0	10.2	9.1
OC	GFDL	9.6	9.4	9.3
	GISS	8.7	8.5	8.0
Sulfate	GFDL	8.0	8.2	8.1
	GISS	8.8	8.8	9.0

The residence times of black and organic carbon (BC and OC) are also fairly similar in these two models (Table 3.3). While the concentrations of sulfate and carbonaceous aerosols are all influenced by differences in how the models simulate removal by the hydrologic cycle, accounting for at least some of the 10-15% difference in residence times, sulfate production can vary even more from model to model, as its production from the emitted sulfur dioxide involves chemical oxidation, which can differ substantially between models. Removal of sulfur dioxide prior to conversion to sulfate may also be more efficient in the GISS model. In contrast, BC and OC are emitted directly, and hence any differences in how these are represented in the models would be apparent in their residence times.

**Table 3.4 Ratio of sulfate and ozone burdens to precursor emissions, global mean annual average**

Species	Model	2000 Tg burden/ Tg emission	2030 vs. 2000 Tg burden/ Tg emission	2030 vs. 2000 % burden/ % emission	2050 vs. 2000 % burden/ % emission
Sulfate	GFDL	0.017	0.017	1.00	0.08*
	GISS	0.012	0.007	0.60	0.65
Ozone	GFDL	7.19	2.24	0.32	0.44
	GISS	6.82	6.54	0.94	0.96

Ratios for sulfate are in Tg sulfate per Tg S/yr SO<sub>2</sub> emitted. Ozone ratios are in Tg ozone per Tg N/yr NO<sub>x</sub> emitted. Ozone values in Table 3.2 are converted to burden assuming 1 DU globally averaged = 10.9 Tg ozone.

\*The burden change was only 2% in this case, making the calculation unreliable.

The aerosol residence times are relatively stable in time in the GISS and GFDL models.

The carbonaceous aerosol residence times do decrease with time in the GISS model (and to a lesser extent in the GFDL model for OC), probably owing to the shift with time from mid to tropical latitudes, where wet and dry removal rates are different (more rapid net removal). The sulfate residence time is fairly stable over the 2000 to 2050 period. The ratio of sulfate burden to SO<sub>2</sub> emissions is the same for the present-day and the 2030 to 2000 changes in the GFDL model. For the 2100 to 2000 change in that model (not shown), the ratio drops from 1.00 to 0.65. As the total emissions of SO<sub>2</sub> decrease, a larger fraction of the sulfate production comes from DMS oxidation rather than from emitted SO<sub>2</sub>. The conversion efficiency from SO<sub>2</sub> to sulfate also varies over time in the GISS model, decreasing to 2030 and increasing thereafter (inversely related to total sulfur dioxide emissions). This may reflect both non-linearities in production (via oxidation chemistry) and the changing spatial pattern of emissions.

After comparison of the inter-model variations in aerosol residence times and chemical conversion efficiencies with the variations in emissions trends, it is clear that the differences in the projected changes in aerosol burdens in the GISS and GFDL

simulations are primarily attributable to the underlying differences in emissions. This is especially true for carbonaceous aerosols, for which the residence times are quite similar in the models. Even though there is a greater difference in sulfate burdens due to the variations in chemical conversion efficiency between the models, the emissions trends at 2050 relative to 2000 are of opposite sign in the two models and thus dominate the difference in the burden change. Thus, the GISS model projects a greater sulfate burden at 2050 than at 2000, but substantially reduced burdens of carbonaceous aerosols, while the GFDL model projects the opposite, both because of the underlying emissions projections.

The results for tropospheric ozone tell a different story. The ozone burden increases in the future in all three models, but the percentage increase relative to 2000 differs by more than a factor of three at 2030 (Table 3.2). Examining the ozone changes relative to the  $\text{NO}_x$  emissions changes, there are very large differences between the GFDL and GISS models (Table 3.4). This may reflect the influence of processes such as stratospheric ozone influx which are independent of  $\text{NO}_x$  emissions, as well as the roles of precursors such as carbon monoxide (CO) and hydrocarbons that also influence tropospheric ozone. In particular, the GISS model computed a large increase in the flux of ozone into the troposphere as the stratospheric ozone layer recovered, while the composition model used at GFDL to calculate ozone held stratospheric ozone fixed and hence did not simulate similar large increases. In addition, there are well-known non-linearities in  $\text{O}_3$ - $\text{NO}_x$  chemistry (Stewart *et al.*, 1977), and it has been shown that the ozone production efficiency can vary substantially with time (Lamarque *et al.*, 2005a; Shindell *et al.*,



2006a). Thus for tropospheric ozone, the differences in modeled changes of nearly a factor of three (13 vs. 33% increase) are much larger than the differences in the NO<sub>x</sub> precursor emissions (33 vs. 43% increase).

### 3.2.4 Aerosol Optical Depth

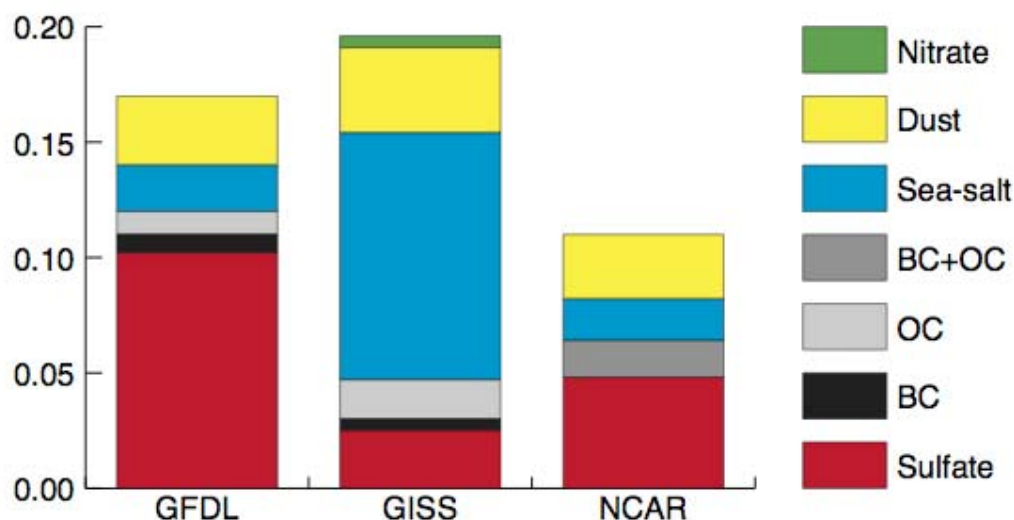
The global mean present-day all-sky aerosol optical depth (AOD)<sup>36</sup> in the three models ranges from 0.12-0.20 (Table 3.5). This difference of almost a factor of 2 suggests that aerosols are contributing quite differently to the Earth's energy balance with space in these models. Observational constraints on the all-sky value are not readily available, as most of the extant measurement techniques are reliable only in clear-sky (cloud-free) conditions. Sampling clear-sky areas only, the GISS model's global total aerosol optical depth is 0.12 for 2000 (0.13 Northern Hemisphere, 0.10 Southern Hemisphere). This includes contributions from sulfate, carbonaceous, nitrate, dust, and sea-salt aerosols. The clear-sky observations give global mean values of ~0.135 (ground-based AERONET) or ~0.15 (satellite composites, including AVHRR or MODIS observations), though these have substantial limitations in their spatial and temporal coverage. The NCAR and GFDL models did not calculate clear-sky aerosol optical depth. Given that the all-sky values are larger, and substantially so in the GISS model (though this will depend upon the water uptake of aerosols), it seems clear that the values for NCAR would be too small compared with observations since even their all-sky values are lower than the estimate from observations. This may be related to NCAR's use of AVHRR data in assimilation of

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<sup>36</sup> Aerosol optical depth is a measure of the fraction of radiation at a given wavelength absorbed or scattered by aerosols while passing through the atmosphere.

aerosol optical depth to create the NCAR climatology (Collins *et al.*, 2001, 2006), as that data appears to be low relative to MODIS observations, for example.

For all three models, there are large differences in the contributions of the various aerosol species (Figure 3.2, Table 3.5). This is true even for GFDL and GISS models, with relatively similar all-sky global mean aerosol optical depths. More than half the aerosol optical depth in the GFDL model comes from sulfate, while this species contributes only about 1/8th the aerosol optical depth in the GISS model. Instead, the GISS model's aerosol optical depth is dominated by the largely natural sea-salt and dust aerosols, which together contribute 0.14 to the aerosol optical depth. These two species contribute a much smaller aerosol optical depth in the NCAR and GFDL models, ~0.06 or less, with the differences with respect to GISS predominantly due to sea-salt. The relative contribution from sulfate in the NCAR model looks similar to the GFDL model, with nearly half its aerosol optical depth coming from sulfate, but the magnitude is much smaller. It seems clear that the GFDL model's direct sulfate contribution is biased high (Ginoux *et al.*, 2006), while the GISS model's sulfate is biased low in this model version (Shindell *et al.*, 2007). However, the relative importance of the different aerosols species is not well understood at present (Kinne *et al.*, 2006).



**Figure 3.2** Present-day contributions from individual aerosol species to global mean all-sky aerosol optical depth (550 nm extinction). Neither GFDL nor NCAR include nitrate.

Large differences in the relative aerosol optical depth in the Northern Hemisphere and Southern Hemisphere are also apparent in the models (Table 3.5). The ratios of the present-day Northern Hemisphere to Southern Hemisphere total aerosol optical depths in the three models differ widely, with values of 2.43, 1.97, and 0.96 in the GFDL, NCAR, and GISS models, respectively. This clearly reflects the dominant contribution of sulfate to optical depth in the GFDL and NCAR models, as this species has large anthropogenic Northern Hemisphere sources, and the dominance of sea-salt in the GISS model, with its largest source being the Southern Ocean. While composite satellite data shows clearly greater aerosol optical depths in the Northern Hemisphere than the Southern Hemisphere, most satellite instruments lose coverage near the northern edge of the Southern Ocean (Kinne *et al.*, 2006). Unfortunately, quality-controlled networks such as AERONET provide virtually no ground-based data poleward of 45°S. Thus while it seems unlikely that the aerosol optical depth is larger in the Southern Hemisphere than the Northern Hemisphere, as in the GISS model, presently available data are not adequate to fully

2046 characterize this ratio, as aerosol optical depths over the Southern Ocean are poorly  
2047 known.

2048

**Table 3.5 Aerosol optical depth (550nm extinction) – ALL-SKY**

Region	aerosol type	Model	2000	2030	2050	2100
Global	BC	GFDL	.0076	.0096	.0105	.0138
		GISS	.0045	.0034	.0028	NA
	Sulfate	GFDL	.1018	.1227	.0906	.0591
		GISS	.0250	.0312	.0278	NA
		NCAR	.048	.062	.052	NA
	Sea-salt	GFDL	.0236	.0236	.0236	.0236
		GISS	.1065	.1080	.1050	
		NCAR	.018	.018	.018	NA
	Dust	GFDL	.0281	.0281	.0281	.0281
		GISS	.0372	.0389	.0387	
		NCAR	.0275	.0275	.0275	NA
	OC	GFDL	.0104	.0122	.0131	.0166
		GISS	.0166	.0135	.0130	
	Nitrate	GISS	.0054	.0057	.0060	
	<b>Total</b>	<b>GFDL</b>	<b>.1715</b>	<b>.1964</b>	<b>.1660</b>	<b>.1411</b>
		<b>GISS</b>	<b>.1959</b>	<b>.2007</b>	<b>.1934</b>	<b>NA</b>
		<b>NCAR</b>	<b>.116</b>	<b>.1392</b>	<b>.1206</b>	<b>NA</b>
Northern Hemisphere	BC	GFDL	.0109	.0147	.0161	.0209
		GISS	.0062	.0043	.0032	NA
	Sulfate	GFDL	.1509	.1766	.1038	.0694
		GISS	.0352	.0449	.0388	NA
		NCAR	.078**	.097**	.073**	NA
	Dust	GISS	.0600	.0642	.0615	
		GFDL	.0491	.0491	.0491	.0491
	Sea-salt	GISS	.0630	.0619	.0647	
		GFDL	.0181	.0181	.0181	.0181
	<b>Total</b>	<b>GFDL</b>	<b>.2430</b>	<b>.2756</b>	<b>.2056</b>	<b>.1807</b>
		<b>GISS</b>	<b>.1910</b>	<b>.1985</b>	<b>.1907</b>	<b>NA</b>
		<b>NCAR</b>	<b>.1538</b>	<b>.1827</b>	<b>.1502</b>	<b>NA</b>
Southern Hemisphere	BC	GFDL	.0042	.0046	.0049	.0066
		GISS	.0029	.0026	.0023	NA
	Sulfate	GFDL	.0526	.0689	.0774	.0487
		GISS	.0148	.0175	.0170	NA
		NCAR	0.052**	.062**	.075**	
	Dust	GISS	.0144	.0137	.0159	
		GFDL	.0071	.0071	.0071	.0071
	Sea-salt	GISS	.1502	.1541	.1453	
		GFDL	.0291	.0291	.0291	.0291
	<b>Total</b>	<b>GFDL</b>	<b>.1000</b>	<b>.1171</b>	<b>.1263</b>	<b>.1015</b>
		<b>GISS</b>	<b>.1997</b>	<b>.2030</b>	<b>.1962</b>	<b>NA</b>
		<b>NCAR</b>	<b>.0779</b>	<b>.0957</b>	<b>.0910</b>	<b>NA</b>

\*\* Total for sulfate + sea salt.

2049

2050

### 3.3 Climate Studies

#### 3.3.1 Experimental Design

The climate studies discussed here consisted of transient climate simulations that were designed to isolate the climate effects of projected changes in the short-lived species and calculate their importance relative to that of the long-lived well-mixed greenhouse gases. The simulations from the GFDL, GISS, and NCAR groups each employed ensembles (multiple simulations differing only in their initial conditions) in order to reduce the unforced variability in the chaotic climate system. One three-member ensemble included the evolution of short- and long-lived species following the A1B scenario, while the second ensemble included only the evolution of long-lived species with the short-lived species fixed at present values. While all three groups used the same values for the long-lived species, each had its own version of an A1B scenario for short-lived species, as discussed previously in Section 3.2.

The global three-dimensional distributions of short-lived gases and aerosols were modeled using each group's chemistry-aerosol composition model. For the first ensemble, the GFDL simulations used aerosol and ozone distributions computed each decade out to 2100, while the GISS and NCAR simulations employed values computed for 2000, 2030, and 2050. Either seasonally varying or monthly-average three-dimensional distributions were saved. Short-lived species concentrations for intermediate years were linearly interpolated between the values for computed years. In both sets of simulations, the concentrations of long-lived species varied with time. In practice, NCAR

performed only a single pair of simulations out to 2050, while GISS performed all three pairs out to 2050, and GFDL extended all three pairs out to 2100.

### 3.3.2 Climate Models

#### 3.3.2.1 Geophysical Fluid Dynamics Laboratory (GFDL)

Climate simulations at GFDL used the comprehensive climate model (Atmosphere-Ocean General Circulation Model (AOGCM); see Box 1.1) recently developed at NOAA's Geophysical Fluid Dynamics Laboratory, which has described in detail in (Delworth *et al.*, 2006). The control simulation of this AOGCM (using present-day values of radiatively active species) has a stable, realistic climate when integrated over multiple centuries. The model is able to capture the main features of the global evolution of observed surface temperature for the 20th century as well as many continental-scale features (Knutson *et al.*, 2006). Its equilibrium climate sensitivity to a doubling of CO<sub>2</sub> is 3.4°C<sup>37</sup> (Stouffer *et al.*, 2006). The model includes the radiative effects of well-mixed gases and ozone on the climate as well as the direct effects of aerosols, but does not include the indirect aerosol effects (see Box 3.1). Further details on the model resolution, model physics, and model performance are included in Appendix 3.2 (Climate Models) in the section on Geophysical Fluid Dynamics Laboratory.

#### 3.3.2.2 Goddard Institute for Space Studies (GISS)

The GISS climate simulations were performed using GISS ModelE (Schmidt *et al.*, 2006). This model has been extensively evaluated against observations (Schmidt *et al.*,

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<sup>37</sup> Equilibrium climate sensitivity is defined here as the global -mean annual- mean surface temperature change of a climate model in response to a doubling of atmospheric carbon dioxide from preindustrial levels, when the model has fully adjusted to the change in carbon dioxide.

2096 2006), and has a climate sensitivity in accord with values inferred from paleoclimate data  
2097 and similar to that of mainstream General Circulation Models.; the equilibrium climate  
2098 sensitivity for doubled CO<sub>2</sub> is 2.6°C. The radiatively active species in the model include  
2099 well-mixed gases, ozone, and aerosols. The model includes a simple parameterization for  
2100 the aerosol indirect effect (Menon *et al.*, 2002) (see box on aerosol indirect effect).  
2101 Further details on the model resolution and model physics are included in Appendix 3.2  
2102 (Climate Models) in the section on Goddard Institute for Space Studies.

### 2104 3.3.2.3 National Center for Atmospheric Research (NCAR)

2105 The transient climate simulations use the NCAR Community Climate System Model  
2106 CCSM3 (Collins *et al.*, 2006). The equilibrium climate sensitivity of this model to  
2107 doubled CO<sub>2</sub> is 2.7°C. Further details on the model resolution and construction are found  
2108 in Appendix 3.2 (Climate Models) in the section on National Center for Atmospheric  
2109 Research.



**Box 3.1: Radiative Effects of Aerosols**

The direct effects of aerosols refer to their scattering and absorption of both incoming solar and outgoing terrestrial radiation. By reflecting incoming radiation back to space, most aerosols have a negative radiative forcing (cooling effect). For reflective aerosols (sulfate, organic carbon, nitrate, dust and sea-salt), this effect dominates over their absorption of outgoing radiation (the greenhouse effect) on the global scale. The balance varies both geographically and seasonally as a function of solar radiation and the ground temperature. In contrast, absorbing aerosols such as black carbon have a positive radiative forcing (warming effect) as they absorb incoming and outgoing radiation, reducing the overall fraction of the sun's irradiance that it reflected back to space. They can also absorb outgoing radiation from the Earth (the greenhouse effect).

In addition to their direct radiative effects, aerosols may also lead to an indirect radiative forcing of the climate system through their effect on clouds. Two aerosol indirect effects are identified: The first indirect effect (also known as the cloud albedo effect) occurs when an increase in aerosols causes an increase in cloud droplet concentration and a decrease in droplet size for fixed liquid water content (Twomey, 1974). Having more, smaller drops increases the cloud albedo (reflectivity). The second indirect effect (also known as the cloud lifetime effect) occurs when the reduction in cloud droplet size affects the precipitation efficiency, tending to increase the liquid water content, the cloud lifetime (Albrecht, 1989), and the cloud thickness (Pincus and Baker, 1994). As the clouds last longer, this leads to an increase in cloud cover. It has been argued that empirical data suggest that the second indirect effect is the dominant process (Hansen *et al.*, 2005).

The direct effects of aerosols are relatively well-represented in climate models such as those described in Section 3.3.2 and used in this study, though substantial uncertainties exist regarding the optical properties of some aerosol types and especially of aerosol mixtures. Because of the inherent complexity of the aerosol indirect effect, climate model studies dealing with its quantification necessarily include an important level of simplification. While this represents a legitimate approach, it should be clear that the climate model estimates of the aerosol indirect effect are very uncertain.

The studies discussed in chapter 3 of this report include the direct effects of aerosols in all three models (though nitrate is only included in the GISS model). The indirect effect is only included in the GISS model, which uses a highly simplified representation of the second indirect effect.

2111

2112 **3.3.3 Radiative Forcing Calculations**

2113 The radiative forcing at the tropopause provides a useful, though limited, indicator of the  
2114 climate response to perturbations (Hansen *et al.*, 2005) (see Box 3.2).

2115

2116

**Box 3.2: Radiative Forcing**

**Radiative forcing** is defined as the change in net (down minus up) irradiance (solar plus longwave, in  $\text{W m}^{-2}$ ) at the tropopause due to a perturbation after allowing for stratospheric temperatures to adjust to radiative equilibrium, but with surface and tropospheric temperatures and state held fixed at the unperturbed values (IPCC, 2007; Ramaswamy et al, 2001). This quantity is also sometimes termed adjusted radiative forcing. If the stratospheric temperatures are not allowed to adjust, the irradiance change is termed instantaneous radiative forcing.

The utility of the radiative forcing concept is that, to first order, the equilibrium global-mean, annual-mean surface temperature change is proportional to the radiative forcing, for a wide range of radiative perturbations (WMO, 1986). The proportionality constant (often denoted as the climate sensitivity parameter,  $\lambda$ ) is approximately the same (to within 25%) for most drivers of climate change (IPCC, 2007), with a typical value of ~0.5-0.7 for most models. This enables a readily calculable and comparable measure of the climate response to radiative perturbations, such as those discussed in this Chapter.

2117

2118 **3.3.3.1 Global and Hemispheric Average Values: GFDL and GISS**

2119 Radiative forcing calculations were performed by GFDL (adjusted forcing) and GISS  
 2120 (instantaneous forcing), but were not performed for the NCAR model. The annual-  
 2121 average global-mean radiative forcing (RF) from short-lived species at 2030 relative to  
 2122 2000 is small in both the GFDL and GISS models (Figure 3.3; Table 3.6). However, this  
 2123 is for quite different reasons. In the GFDL model, a large increase in sulfate optical depth  
 2124 leads to a negative forcing that is largely balanced by positive forcings from increased  
 2125 black carbon aerosol and ozone. In the GISS model, increased sulfate and reduced black  
 2126 carbon both lead to relatively small negative forcings that largely offset a substantial  
 2127 positive forcing from increased ozone. Moving to 2050, the models now diverge in their  
 2128 net values as well as the individual contributions. The GFDL model finds a positive  
 2129 radiative forcing due in nearly equal parts to increased black carbon and ozone. In  
 2130 contrast, 2050 radiative forcing in the GISS model again reflects an offset between  
 2131 positive forcing from ozone and negative aerosol forcing, with the largest contribution to

the latter from reduced levels of black carbon. Both models show a partial cancellation of the black carbon forcing by an opposing forcing from organic carbon. Thus, the two models show somewhat consistent results for ozone, but differ dramatically for black carbon and sulfate aerosol. By 2100, the GFDL model has a large positive radiative forcing relative to 2000, due to the continued increase in black carbon as well as the decrease in sulfate.

**Table 3.6 Global mean radiative forcing for short-lived species ( $\text{W m}^{-2}$ )**

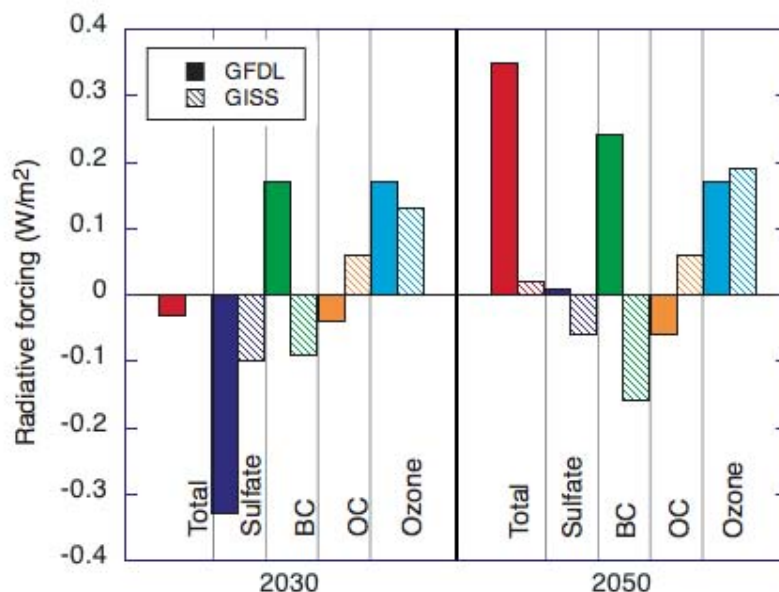
	Model	2030	2050	2100
Total	GFDL	.04	.48	1.17
	GISS	.00	.02	NA
Aerosols	GFDL	-.15	.24	.98
	GISS	-.13	-.17	NA
Sulfate	GFDL	-.32	.01	.51
	GISS	-.10	-.06	NA
BC	GFDL	.21	.30	.63
	GISS	-.09	-.16	NA
OC	GFDL	-.04	-.06	-.15
	GISS	.06	.06	NA
Ozone	GFDL	.19	.23	.19
	GISS	.13	.19	NA

Values are annual average radiative forcings at the tropopause (meteorological tropopause in the GISS model, 'linear' tropopause in the GFDL model). 'Aerosols' is the total of sulfate, black carbon (BC), and organic carbon (OC) (plus nitrate for GISS). GISS values do not include aerosol indirect effects that were present in that model.

GFDL values are for adjusted radiative forcing; GISS values are for instantaneous radiative forcing (see Box 3.2). The GFDL values are from (Levy *et al.*, 2007).

Inter-model differences in radiative forcing are predominantly due to differences in modeled burdens rather than to differences in the calculation of radiative properties in the models. This can be seen clearly by examining the RF-to-burden ratio, which we term the radiative efficiency (Table 3.7). This shows fairly similar values for GFDL and GISS. The largest differences are seen for black carbon, which may reflect differences in the geographic location of projected black carbon changes as well as differing treatments of

2153 the radiative properties of black carbon. Additionally, the vertical distribution of the  
2154 black carbon changes will affect the radiative forcing, as will their location relative to  
2155 clouds. Variations in modeling the aerosol uptake of water, which can have a substantial  
2156 impact on the aerosol optical depth , do not seem to play a very large role in the global  
2157 mean radiative forcing judging from the fairly close agreement in the two models' sulfate  
2158 radiative efficiencies (Table 3.7). They may contribute to the ~20% difference in the RF-  
2159 to-burden ratios for sulfate, however. Examination of the RF-to-aerosol optical depth  
2160 change (Table 3.7) shows that given a particular aerosol optical depth change, the models  
2161 are in good agreement as to the resulting radiative forcing. We caution that this result  
2162 contrasts with a wider model study that found larger differences in this ratio (Schulz *et*  
2163 *al.*, 2006), though the variation in RF-to-aerosol optical depth across models was still  
2164 less than the variation in aerosol optical depth itself. This suggests a possible further  
2165 source of model differences that could exist were different models to be used in a study  
2166 such as this.



**Figure 3.3** Global mean annual average radiative forcing (in  $\text{W m}^{-2}$ ) from short-lived species at 2030 and 2050 relative to 2000. Values from the GFDL model are shown as solid bars; values from the GISS model have diagonal hatching. (Note that instantaneous forcing values from the GFDL model are shown in this figure, not the adjusted forcings shown in Table 3.6.)

Both the GFDL and GISS models show a positive forcing from ozone that stems partially from increased tropospheric ozone concentrations (Table 3.2) due to increased  $\text{NO}_x$  emissions (Table 3.1) and partially from the recovery of stratospheric ozone due to reductions in emissions of ozone-depleting substances (primarily halogens). The forcing from the tropospheric portion of the ozone changes is substantially more important, however (Shindell *et al.*, 2007). (The NCAR group did not calculate the radiative forcing, but forcing in their model is likely to have been similar, as they found an increase in the tropospheric ozone burden from 2000 to 2050 of 15.0 Dobson Units (DU), very close to the GISS value of 16.2 DU (Table 3.2).) As shown previously, however, the apparent sensitivity of ozone burden to changes in  $\text{NO}_x$  emissions differs substantially between the GISS and GFDL models. Thus the similarity in the radiative forcing may be largely

fortuitous, resulting from a cancellation of changes in emissions and of sensitivities of ozone to NO<sub>x</sub> emissions.

Thus, at 2030, differences in the physical processes in the two models dominate the differences in radiative forcing between the two models. The large divergence in radiative forcing from sulfate stems from both the chemical conversion efficiency of SO<sub>2</sub> to sulfate being more than a factor of two larger in the GFDL model than in the GISS model, and the greater role of sulfate in producing aerosol optical depth in the GFDL model. In addition, the GISS model includes a substantial absorption of sulfate onto dust, a process that is highly uncertain. Such a process would reduce the radiative forcing due to sulfate. At 2050, emissions and concentrations of sulfur dioxide have returned to near their 2000 level, so that these differences are not so important at this time. Hence, the 2050 differences between the two models are dominated by differences in black carbon emissions projections and not by differences in physical processes. Differences in the residence times and radiative efficiencies for black carbon are substantial but tend to offset.

**Table 3.7 Radiative efficiency.**

Species	Model	(W m <sup>-2</sup> )/Tg	(W m <sup>-2</sup> )/AOD
BC	GFDL	2.8	104
	GISS	1.5	94
OC	GFDL	-0.18	NA
	GISS	-0.16	NA
Sulfate	GFDL	-.47	-16
	GISS	-.59	-16

Values are given for the radiative efficiency in terms of the RF-to-burden ratio and the RF-to-aerosol optical depth (AOD) ratio. All values are global-mean, annual-mean averages. Values for radiative forcing and burden or aerosol optical depth changes are for 2050 versus 2000 for black carbon (BC) and organic carbon (OC), and 2030 versus 2000 for sulfate in order to analyze the largest changes for each species. GISS values for the sulfate burden changes include only the portion of sulfate not absorbed onto dust, as this portion alone is radiatively important.

On a hemispheric scale, the GISS and GFDL models again differ greatly (Table 3.8). The GFDL model shows a very large positive forcing in the Northern Hemisphere in 2050 due primarily to reductions in emissions of sulfate precursors and increased emissions of black carbon. Increases in sulfate precursor emissions from developing countries lead to a negative forcing in the Southern Hemisphere in the GFDL model. In the GISS model, the sign of the total net forcings are reversed, with negative values in the Northern Hemisphere and positive in the Southern Hemisphere (Table 3.8). The GISS results are primarily due to the reduction in BC in the Northern Hemisphere and the influence of increased ozone and reduced OC in the Southern Hemisphere (where sea-salt dominates the aerosol optical depth, so that anthropogenic aerosol emissions changes are relatively less important).

**Table 3.8 Hemispheric radiative forcing ( $\text{W m}^{-2}$ ).**

	Model	2030	2050	2100
Northern Hemisphere	GFDL	.15	1.09	1.91
	GISS	-.15	-.14	NA
Southern Hemisphere	GFDL	-.09	-.14	.42
	GISS	.16	.18	NA

Values are the net annual average forcings at the tropopause in each hemisphere from aerosols and ozone. GISS forcing values do not include aerosol indirect effects that were present in that model.

### 3.3.3.2 Regional Forcing Patterns

The differences in hemispheric and global forcings can be attributed to strong forcing changes in particular regions, and hence to regional emissions as the radiative forcing is typically localized relatively close to the region of emissions. Comparison of the spatial patterns of radiative forcing in the GISS and GFDL models reveals that the starkest discrepancies occur in the developing nations of South and East Asia (Figure 3.4). The

emissions scenario used by the GISS model projects strong increases in SO<sub>2</sub> emissions from India, with little change over China. In contrast, the scenario used by the GFDL model has large decreases in sulfate emissions in both regions, especially China.

The scenarios are much more similar for the developed world, with both projecting reductions in sulfate precursor emissions for North America and Europe, for example, leading to a positive radiative forcing in both cases. Differences between the scenarios are even larger for black carbon, which increases throughout most of the Northern Hemisphere in the GFDL model but decreases in the GISS model. Again, however, the divergence is especially large over South and East Asia, where the GISS model has large reductions while the GFDL model has large increases (Figure 3.4). Thus the differences in the global total emissions discussed previously (Figure 3.1; Table 3.1) and in the global radiative forcing (Figure 3.2; Table 3.6) arise primarily from differences in projected emissions from developing countries in Asia.

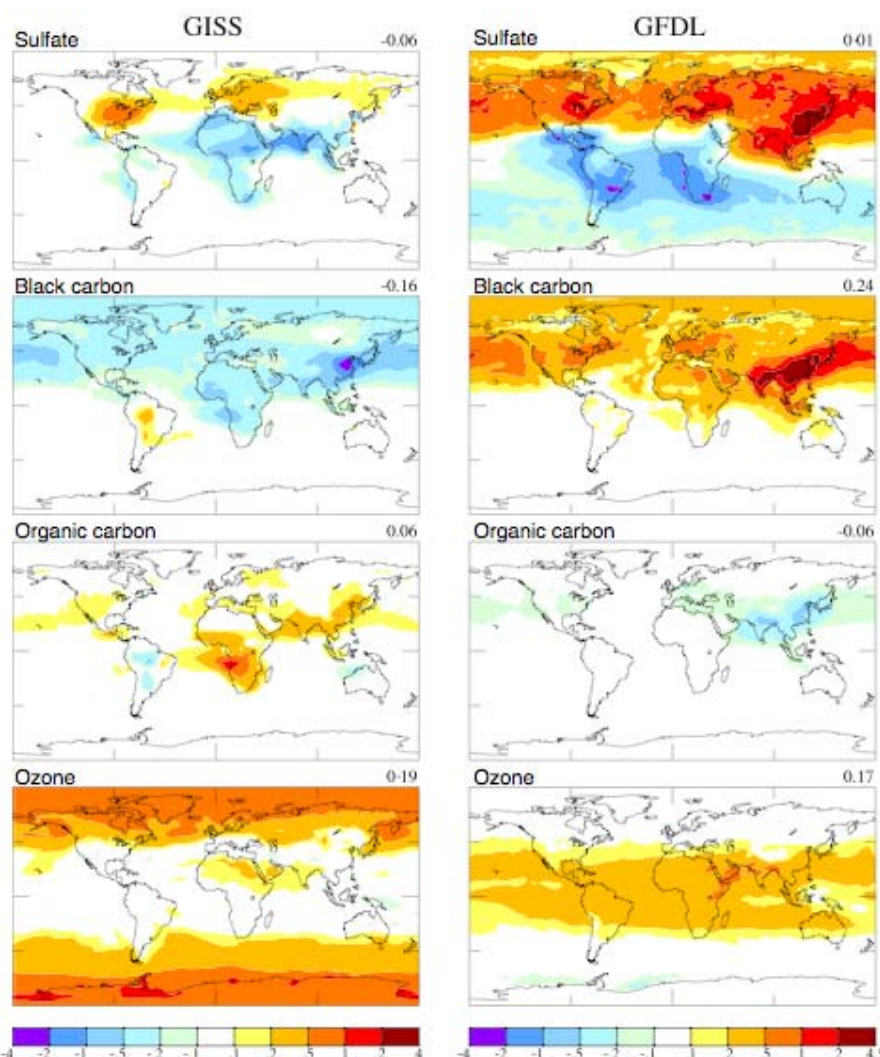
The radiative forcing from organic carbon is generally similar in its spatial pattern to black carbon, but of opposite sign and substantially reduced magnitude (25-40% of the black carbon radiative forcing). Substantial differences again occur between the emissions scenarios of the two models, this time primarily over African biomass burning regions. As discussed previously, the GFDL model assumed that biomass burning emissions would scale with ½ the factor used for purely anthropogenic emissions, while the GISS model instead used regional biomass burning emissions projections from (Streets *et al.*, 2004), with substantial reductions in African biomass burning.



2255

2256 The spatial pattern of radiative forcing from ozone is also very different in the two  
2257 models (Figure 3.4). However, this forcing is not so closely tied to the region of precursor  
2258 emissions in the GISS model where much of the forcing is related to an increased flux of  
2259 ozone into the troposphere owing to the recovery of lower stratospheric ozone. This leads  
2260 to substantial positive forcing in that model at high latitudes, even without including the  
2261 effects of climate change on circulation (see section 3.3.3.4). At low latitudes, GISS  
2262 shows little forcing as the modeled increase in upper stratospheric ozone causes cooling  
2263 which offsets part of the warming caused by lower stratospheric ozone increases.  
2264 Additionally, the overall increase in overhead stratospheric ozone reduces the  
2265 photochemical formation of ozone in the troposphere, offsetting some of the  
2266 enhancement there due to increased precursor emissions. The GFDL model does not  
2267 show a similar high latitude enhancement, however, but instead shows maximum ozone  
2268 forcing in the tropics. This may reflect a greater geographic shift in emissions to lower  
2269 latitudes, a greater efficiency in transporting ozone and its precursors to the upper  
2270 troposphere, where ozone has the greatest positive forcing efficiency, and differences in  
2271 the relative importance of change in the overlying stratospheric ozone column. The  
2272 GFDL radiative forcing is similar to results from models with tropospheric ozone only  
2273 (Gauss *et al.*, 2003).

2274



**Figure 3.4** Annual average instantaneous radiative forcing ( $\text{W m}^{-2}$ ) near 2050 relative to 2000 for the indicated individual short-lived species in the GISS (left) and GFDL (right) models. Radiative forcing from long-lived species is largely spatially uniform over the globe. (Note that the instantaneous forcings shown here for the GFDL model differ from the adjusted forcings shown in Table 3.6.).

### 3.3.3.3 Effects of uncertainties in methane concentrations on radiative forcing

The SAP 3.2 simulations included methane concentrations prescribed to A1B values from the AIM integrated assessment model, for consistency with the long-lived species runs. To investigate the potential uncertainty in the methane value derived by that integrated assessment model, the GISS model performed an additional 2050 simulation using its

internal methane cycle model (Shindell *et al.*, 2007). The simulation included prescribed anthropogenic emissions increases from the AIM model to allow comparisons with the AIM results used in the results in this report (SAP 3.2). Natural spatially and seasonally varying emissions and soil adsorption were the standard amounts described in (Shindell *et al.*, 2003). Both the methane emissions from wetlands and the biogenic isoprene emissions were interactive with the climate in this run (Guenther *et al.*, 1995; Shindell *et al.*, 2004), though the distribution of vegetation did not respond to climate change.

Methane's oxidation rate is calculated by the model's chemistry scheme in both the troposphere and stratosphere. Thus methane can affect its own lifetime (which is primarily governed by tropospheric oxidation rates), as can other molecules that compete with methane for hydroxyl radicals (the main oxidizing agent), such as isoprene. The simulations included 2050 surface climate (sea-surface temperatures and sea ice, taken from an earlier climate model run). Changes in water vapor induced by the altered climate affect methane oxidation in those runs. Methane was initialized with estimated 2050 abundances and the simulations were run for 3 years. We note that the IMAGE integrated assessment model projected a continuous increase in methane emissions, rather different from the increase through 2030 and slow decrease thereafter in the AIM integrated assessment model. At 2050, for example, this led to projected anthropogenic methane emissions of 512 Tg/yr in the IMAGE model, substantially greater than the 452 Tg/yr from the AIM model used here (compared with 323 Tg/yr for 2000).

We find that methane emissions from wetlands increase from 195 to 241 Tg/yr while emissions of isoprene increase from 356 to 555 Tg/yr. Additionally, even in the absence of changes in emissions from natural sources, the projected anthropogenic emissions of ozone precursors (including methane itself) increase the lifetime of methane while climate change reduces it via increased temperature and water vapor (Table 3.9). These responses to anthropogenic emissions and to climate change without interactive emissions are qualitatively consistent with those reported from a range of models (using different emissions projections) in (Stevenson *et al.*, 2006). The effect of precursor emissions is stronger in our scenario, so that the net effect of anthropogenic emissions and climate changes is to increase methane's lifetime. When natural emissions are also allowed to respond to climate change, increased competition from isoprene and increased methane emissions from wetlands lead to further increases in methane's lifetime (Table 3.9) and enhanced methane abundance.

The 2050 simulation with the model's internal methane cycle had a global mean surface methane value of 2.86 ppmv in year 3, with sources exceeding sinks by 80 Tg/yr (a growth rate that may reflect an overestimate of the loss rate in the AIM model used in the initial guess). Extrapolating the change in methane out to equilibrium using an exponential fit to the three years of model results yields a 2050 value of 3.21 ppmv.

We have calculated radiative forcings using the standard calculation (Table 6.2 in (Ramaswamy *et al.*, 2001)) assuming an increase in N<sub>2</sub>O from 316 to 350 ppb in 2050, following the A1B 'marker' scenario (using the AIM integrated assessment model). The

2050 methane forcing using the methane concentration specified in the A1B ‘marker’ scenario would be  $0.22 \text{ W m}^{-2}$  while using the larger methane concentrations of 2.86 or 3.21 ppmv calculated with our model gives 0.36 or  $0.46 \text{ W m}^{-2}$ , respectively. Of course, it is difficult to estimate methane’s abundance at a particular time without performing a full transient methane simulation. However, uncertainty in the forcing from methane appears to be at least  $0.1\text{-}0.2 \text{ W m}^{-2}$ . Note that use of the 40% larger anthropogenic methane emission increase from the IMAGE integrated assessment model would have led to a substantially larger forcing. Should the results of our modeling of the methane cycle prove to be fairly robust, this would imply that future positive forcing from methane might be substantially larger than current estimates based on integrated assessment model projections.

We note that while the A1B projections assume a substantial increase in atmospheric methane in the future, the growth rate of methane has in fact decreased markedly since the early 1990s and leveled off since ~1999 (Dlugokencky *et al.*, 2003). Hence, the projections may overestimate future atmospheric concentrations. However, there are indications that the growth rate decrease was primarily due to reduced anthropogenic emissions, and that these have been increasing again since 1999 (though masked by a coincident decrease in natural methane emissions) (Bousquet *et al.*, 2006). All of this suggests that atmospheric methane may in fact increase substantially again in the future, as assumed by the integrated assessment models, although other methane studies have argued for an increase in its principle loss path as the explanation, rather than changes in emissions (*e.g.* Fiore *et al.*, 2006). Other emissions, such as NO<sub>x</sub> from lightning and from

soil and dimethyl-sulfide from the oceans, are also expected to respond to climate change. Changes in land cover would also affect both emissions and removal of trace species. Further work is required to gauge the importance of these and other climate-chemistry feedbacks.

**Table 3.9** Methane lifetime in GISS simulations. Includes calculated photochemical loss (in troposphere and stratosphere) and prescribed 30 Tg/yr loss to soils.

Run	Lifetime (years)
2000	9.01
2030	9.96
2050	10.39
2030 with climate change	9.72
2050 with climate change	10.01
2050 with methane cycle	10.42

### 3.3.3.4 Effects of Climate Change on Radiative Forcing

The chemical composition simulations (Section 3.2) did not include the effects of climate change on the short-lived species, only the effects of projected changes in anthropogenic emissions. Separate sets of simulations with the GISS model included climate change via prescribed sea-surface temperatures and sea-ice cover taken from prior runs. Climate change increased the radiative forcing from ozone by increasing stratosphere-troposphere exchange (STE) and hence ozone near the tropopause where it is most important radiatively (Hansen *et al.*, 1997). This effect outweighed increased reaction of excited atomic oxygen with the enhanced tropospheric water vapor found in a warmer climate, which led to ozone reductions in the tropical lower troposphere. The overall impact was to increase radiative forcing by  $.07 \text{ W m}^{-2}$  in 2050. Climate change slightly increased the negative forcing from sulfate (by  $.01 \text{ W m}^{-2}$ ), consistent with an increase in tropospheric ozone in these runs (as ozone aids in sulfur dioxide oxidation both directly and via hydroxyl formation).

Dust emissions decreased slightly (~5% at 2050) in these climate runs, but there was more sulfate on dust, suggesting that this played only a minor role in the sulfate forcing response to climate change. The reduction in dust would itself lead to a slight negative forcing ( $\sim 0.02 \text{ W m}^{-2}$ ). However, emissions in the model respond only to changes in surface wind speeds, and not to changes in sources due to either CO<sub>2</sub> fertilization or climate-induced vegetation changes which have a very uncertain effect on future dust emissions (Mahowald and Luo, 2003; Woodward *et al.*, 2005).

Much of the increase in ozone forcing results from an increase in stratosphere-troposphere exchange in the GISS model of 134 Tg/yr (~27% of its present-day value) as climate warms. An increase in transport rates between the stratosphere and the troposphere is a robust projection of climate models (Butchart *et al.*, 2006). Combined with the expected recovery of stratospheric ozone, this should enhance the influx of stratospheric ozone into the lower atmosphere. However, the net effect of climate change on ozone is more difficult to determine as it results from the difference between enhanced stratosphere-troposphere exchange and enhanced chemical loss in the troposphere in a more humid environment, which is not consistent among climate models (Stevenson *et al.*, 2006).

#### 3.3.4 Climate Model Simulations 2000 – 2050

As discussed in Section 3.3.1, the experimental design consists of two sets of simulations:

- 1) the effects of changes in short-lived and long-lived species in the 21<sup>st</sup> century

(employing the A1B scenario for the evolution of the long-lived species and the output from the composition models discussed on Section 3.2 for the short-lived species); 2) the effects of changes in the long-lived species only, with the short-lived species concentrations held at 2000 values. The effects of short-lived species changes are determined by subtracting the climate responses of the runs with changes in long-lived species only from those with changes in long-lived and short-lived species. Simulations where only the short-lived species change were not included in the experimental design, because such a scenarios are neither realistic nor policy relevant

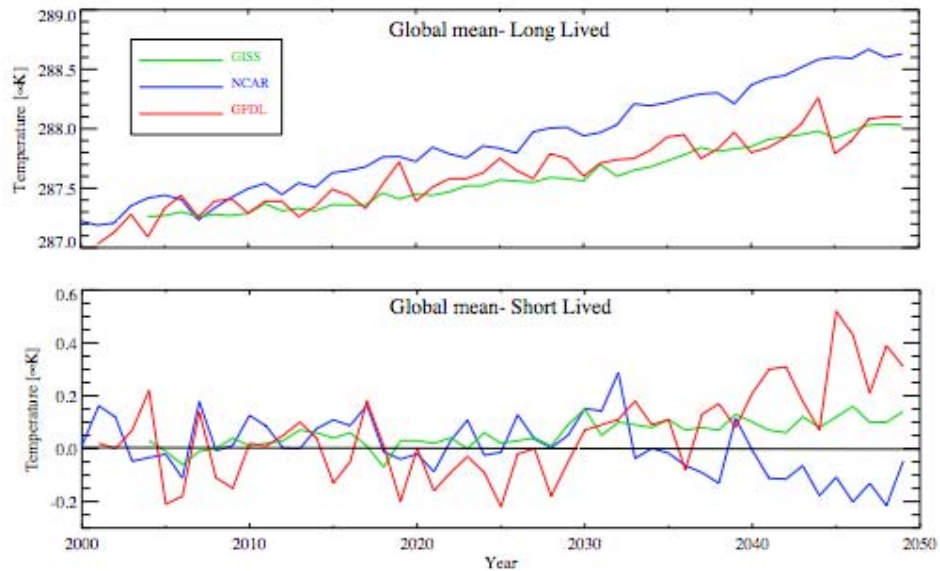
#### 3.3.4.1 Surface Temperature Changes

The global-mean annual-mean surface temperature responses to short-lived species are not as dissimilar as one might have expected, given the different emissions used and the different physical processes included. The NCAR model ran only a single simulation, which showed little or no statistically significant<sup>38</sup> effects of the short-lived species on global mean surface temperatures. The GFDL and GISS models both ran 3-member ensemble simulations, and both show a statistically significant warming effect from short-lived species from around 2030 to the end of the runs (Figure 3.5). The GFDL model shows a warming of 0.28 K (ensemble mean 2046-2050). This value is commensurate with the adjusted radiative forcing of  $\sim 0.48 \text{ W m}^{-2}$ , computed for 2050. The GISS model shows substantially more warming,  $\sim 0.13 \text{ K}$  near 2050, than would be expected from the direct radiative forcing in that model and its climate sensitivity ( $\lambda = \sim 0.6 \text{ K (W m}^{-2})^{-1}$ ) owing to the presence of the aerosol indirect effect, which contributes additional warming as aerosol loading decreases in the future (Shindell *et al.*, 2007).

<sup>38</sup> The statistical methods used to assess significance are discussed in Box 3.3.



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2427

**Figure 3.5** Global mean annual average temperature in the simulations with time-varying long-lived (top) and short-lived (bottom) species. Results are 3-member ensemble means for GFDL and GISS and single-member simulations for NCAR. Results for the short-lived species are obtained by subtraction of the (long-lived) species calculations from the (short + long-lived) species calculations.

2428 The overall global annual average influence of short-lived species is to augment the  
2429 warming from well-mixed greenhouse gases by ~20-25% in these two models (17% for  
2430 GISS and 27% for GFDL based on 2046-2050 vs. the first 5 years of the run). It is  
2431 important to note, however, that these models responded as they did for different reasons.

**Box 3.3: Statistical Methods**

A result is deemed to be statistically significant if it is unlikely to have occurred by chance (*i.e.*, the probability that it occurred by chance is less than some specified threshold). A 95% confidence level means that the odds are 20:1 against the result having occurred by chance..

Statistical significance in the GFDL climate model results was evaluated using two approaches. For global-mean, or hemispheric-mean results involving a temperature departure from the initial (2000) value, the range (highest to lowest temperature change) of the three ensemble members used to obtain the ensemble-mean result was computed. The ensemble-mean result was deemed significant if that range was entirely different from zero. For regional (latitude-longitude) results comprising the difference of two time series, as in the evolution of temperature change due to short-lived species, the student-t test for significance was applied at each model grid point, with the result deemed significant if the statistical test showed significance at the 95% confidence level.

2432  
2433 In the GFDL simulations, reduced sulfate and increased black carbon and ozone all  
2434 combined to cause warming. In contrast, in the GISS model, the warming resulted from  
2435 increased ozone and a reduced aerosol indirect effect, with a substantial offset (cooling)  
2436 from reduced black carbon. The lack of a substantial effect from short-lived species in the  
2437 NCAR simulations is attributable to the emissions used, which produced small increases  
2438 in sulfate (cooling) and small increases in black carbon (warming) that largely offset one  
2439 another (thus their aerosol optical depth changed little from 2000 to 2050).

2440

2441 Hemispheric temperatures show trends largely consistent with the radiative forcings  
2442 (Table 3.8), namely substantial warming in the Northern Hemisphere in the GFDL model  
2443 and in the Southern Hemisphere in the GISS model (Figure 3.6). The Northern  
2444 Hemisphere warming in the GFDL model is driven primarily by the large decreases  
2445 projected for sulfate and the large increase projected for black carbon in that model for  
2446 the industrialized areas of the Northern Hemisphere (Levy *et al.*, 2007). This causes the  
2447 aerosol optical depth from sulfate to drop by 1/3 in the Northern Hemisphere by 2050  
2448 while the aerosol optical depth from black carbon increases by 50%. The large change in

sulfate dominates the overall aerosol optical depth change in that model (Table 3.5). The magnitude of the Northern Hemisphere warming is ~0.5 K by 2050, consistent with the ~1.1 W m<sup>-2</sup> radiative forcing in that model, when one accounts for the fact that the warming has not been fully realized due to the lag-time for oceanic heat adjustment. There is an overall negative forcing in the Southern Hemisphere in the GFDL model, as sulfate precursor emissions increase in the developing world while black carbon changes little. Some of the negative forcing from aerosols in the Southern Hemisphere is offset by positive forcing from ozone, which increases rather uniformly over much of the world in that model (Levy *et al.*, 2007), leading to a small net effect and minimal temperature change from short-lived species (Figure 3.6).

The change in the forcing due to the aerosol indirect effect in the GISS model was argued to be on the order of 0.1 W m<sup>-2</sup> in 2050 (Shindell *et al.*, 2007). Combining this with the GISS hemispheric radiative forcings (excluding the indirect effect) in Table 3.8 yields a Northern Hemisphere radiative forcing near zero and a Southern Hemisphere forcing of ~0.3 W m<sup>-2</sup>. These forcings are consistent with the warming of ~0.15 K seen in that model in the Southern Hemisphere and the lack of response in the Northern Hemisphere. Northern Hemisphere aerosol optical depth changes are dominated by a substantial reduction in black and organic carbon (the black carbon aerosol optical depth in the Northern Hemisphere falls by nearly 50%), which more than offsets a slight increase in sulfate (particularly as this model is less sensitive to sulfate). These aerosol changes lead to negative Northern Hemisphere forcing. In the Southern Hemisphere, the GISS model shows only small changes in aerosols, so that positive forcing from ozone dominates the

net radiative forcing. The aerosol indirect effect further accentuates the positive forcing owing to reductions in black carbon and organic carbon. The signs of the temperature response in the two hemispheres are thus opposite in the GISS model to what they are in the GFDL model.

As for the global case, trends in the NCAR model are not significantly different in the runs with and without short-lived species. This is the result of only a miniscule change in aerosol optical depth in the Northern Hemisphere (-2%), as sulfate and carbonaceous aerosol precursor emissions are both near their present-day values by 2050 in that model. In the Southern Hemisphere, there is an increase in aerosol optical depth from 2000 to 2050, which seems to be primarily due to sulfate, but this is largely offset by increased ozone in the Southern Hemisphere as stratospheric ozone recovers.

Thus it is clear that at global and especially at hemispheric scales, the three climate models are being driven by substantially different trends in their aerosol species. These differences in aerosols are largely related to the differences in the projected emissions of aerosol precursors, though there is some contribution from differences in aerosol modeling as discussed previously. Additionally, the climate response of each model is different to some extent owing to the inclusion of different physical processes in the models, especially the inclusion of the aerosol indirect effect in the GISS model.

However, the above analysis strongly suggests that the largest contributor to the inter-model variations in projected warming arise from different assumptions about emissions trends.

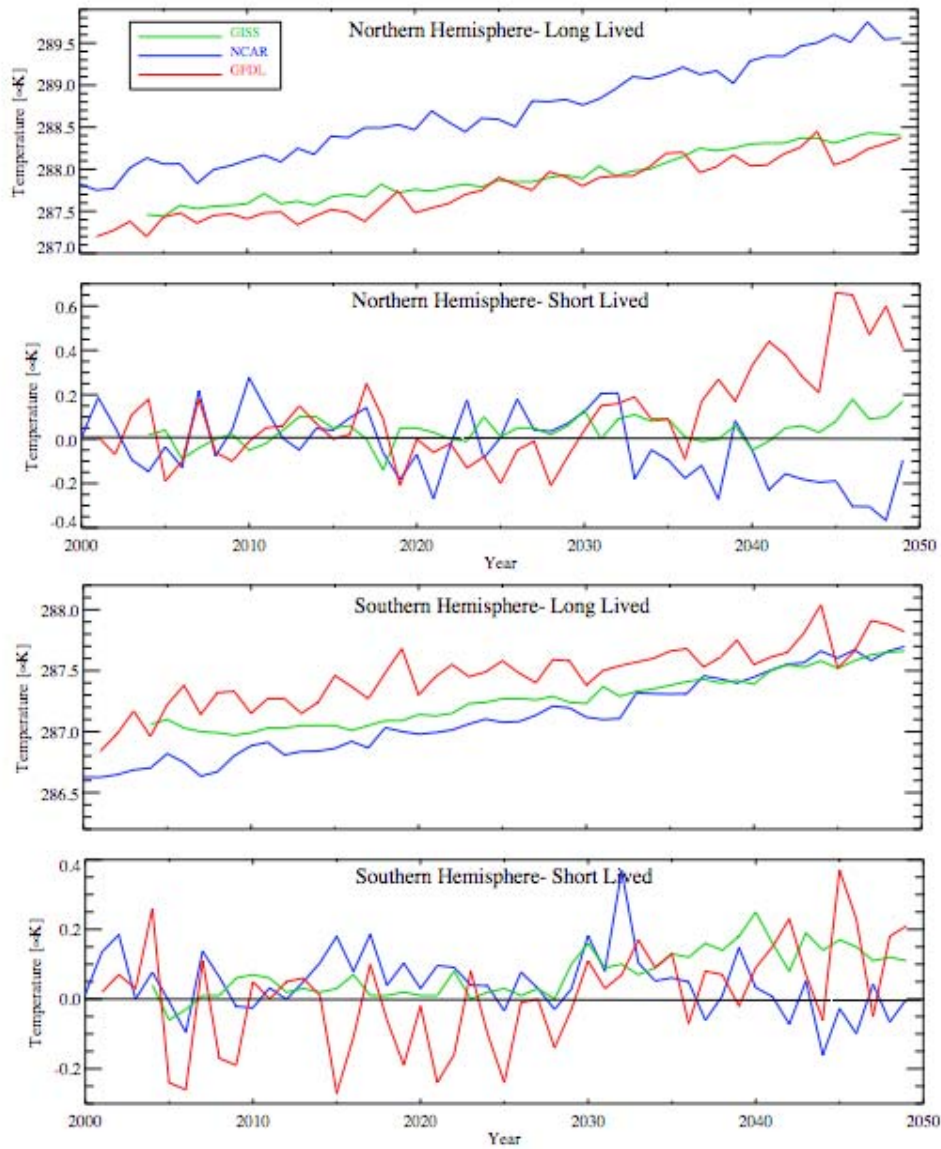
2495

2496 At smaller spatial scales, the annual average patterns of surface temperature changes  
2497 induced by the short-lived species show even larger divergences (Figure 3.7). Around  
2498 2030, the largest responses are seen at Northern middle and high latitudes. These show  
2499 large regions of both cooling and warming that are characteristic of the response to  
2500 dynamic variations. Surprisingly, all three models show cooling near Alaska and a region  
2501 of warming over Siberia. However, most of the response at these latitudes is not  
2502 statistically significant in the models owing to large natural variability. Other regions,  
2503 such as the Labrador Sea/Baffin Island area or Scandinavia, show substantial variations  
2504 between models, again suggesting these middle and high latitude dynamic responses are  
2505 not robust.

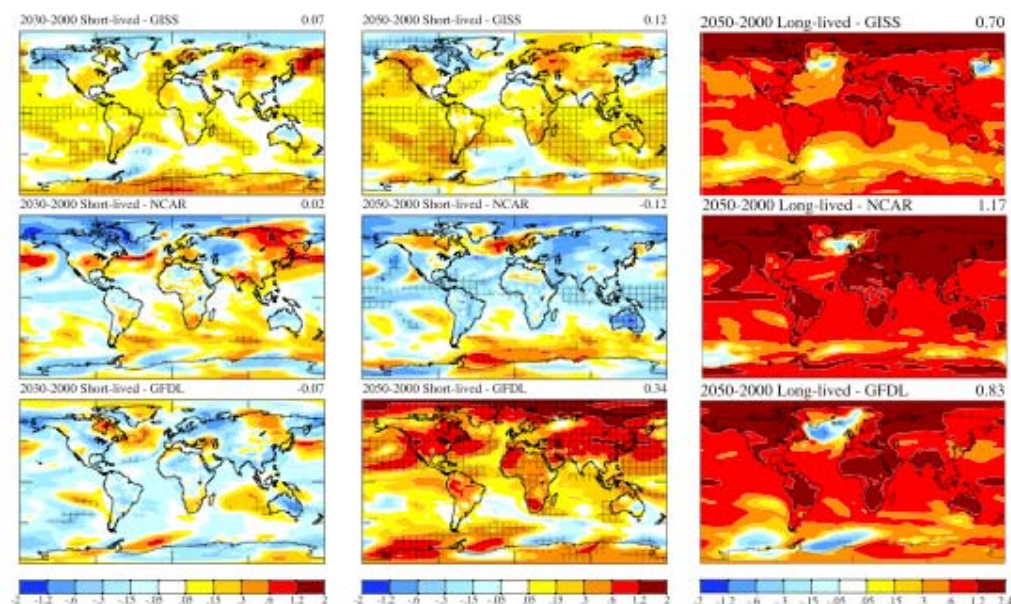
2506

2507 In the tropics, where dynamic variability is much smaller, the models find much greater  
2508 areas with statistically significant responses, especially by 2050. The NCAR model finds  
2509 a small but significant cooling over tropical oceans, while the other models find warming.  
2510 As in the global-mean case, this appears to arise from differences in aerosol burdens and  
2511 optical depths.

2512



**Figure 3.6** Hemispheric mean annual average temperature in the simulations with time-varying long-lived and short-lived species. Results are ensemble means for GFDL and GISS.



**Figure 3.7** Annual average surface temperature response (K) in the climate models to short-lived species (left and center columns) and long-lived species (right column) changes for the indicated times. The changes at 2030 are 2020-2029 in the NCAR and GFDL models and 2028-2033 in the GISS model. At 2050, they are 2040-2049 in the NCAR model, 2046-2055 in the GFDL model, and 2040-2050 in the GISS model. Hatching indicates statistical significance (95%) for the response to short-lived species. Virtually all colored values are statistically significant in the response to long-lived species. Values in the upper right corners give the global mean.

In the Arctic, the GISS and NCAR models find primarily a cooling effect from projected changes in short-lived species (especially near 2030 for GISS, and 2050 for NCAR). In contrast, the GFDL model finds a substantial warming there. This may be due in part to the increasing trend in black carbon in that model.

In the Antarctic, the GISS model shows warming related primarily to stratospheric ozone recovery. The GFDL model shows a similar result by 2050 (after which stratospheric ozone was unchanged in that model). NCAR does not show as clear an Antarctic warming, however, even though this model also included recovery of ozone in the Antarctic lower stratosphere. This is surprising given that the NCAR model appeared to

show a substantial response to ozone depletion in analyses of the Southern Hemisphere circulation in IPCC AR4 simulations (Miller *et al.*, 2006b). That analysis showed that most climate models found a general strengthening of the westerly flow in the Southern Hemisphere in response to stratospheric ozone depletion. A stronger flow isolates the polar region from lower latitude air, leading to cooling over the Antarctic interior and warming at the peninsula. Conversely, recovery should lead to warming of the interior (enhanced by the direct positive radiative forcing from increased ozone), as in the GISS and GFDL simulations. However, in the GISS model the effect diminishes with time, suggesting that other aspects of the response to short-lived species become more important in these scenarios over time, presumably as projected aerosol changes grow ever larger.

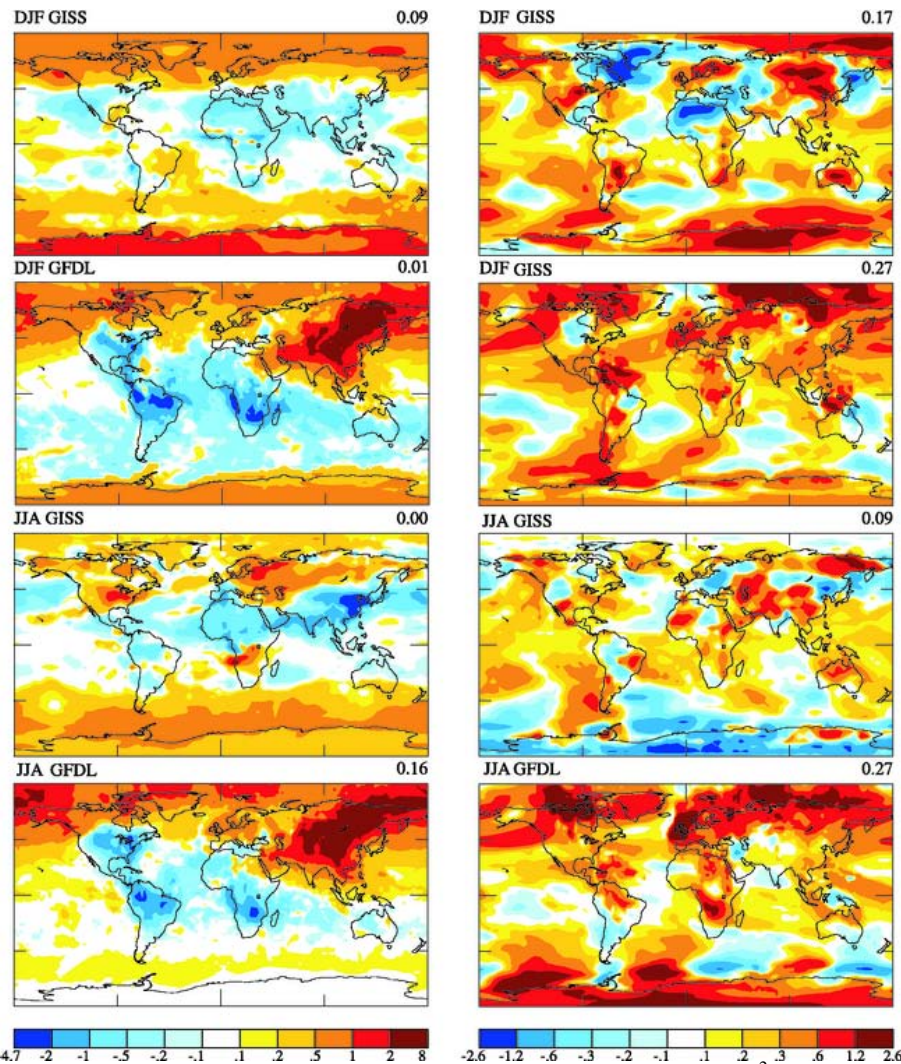
Warming over the central United States is present in the GISS model at all times (but is not statistically significant), in the GFDL model from about the 2040s on, and in the NCAR model around 2030, but not at 2050. The United States and other Northern Hemisphere industrialized regions might be especially sensitive due to the projected reduction in sulfate precursor emissions in the Northern Hemisphere. This effect is especially large in the GFDL model, where forcings from sulfate decreases and black carbon increases both contribute to warming, though it should be noted that the largest radiative forcing is over Asia, not over the US and Europe (Fig. 3.4). In the NCAR model, the warming effect vanishes by 2050 as both sulfate and black carbon decrease, producing temperature responses of opposite sign. In the GISS model, reductions in



sulfate and increases in ozone both contribute to warming; however, these are partially offset by cooling from reduced black carbon.

The surface temperature changes induced by the long-lived species are clearly much larger than those induced by short-lived species over most of the Earth by 2050 (Figure 3.7). In some regions, however, the two are of comparable magnitude (*e.g.* the polar regions and parts of the Northern mid-latitude continents in the GFDL model, parts of the Southern Ocean in the GISS model). Consistency between the models is also clearly greater in their response to long-lived than to short-lived species.

Overall, it is clear that the regional surface temperature response does not closely follow the regional radiative forcing patterns based on either GISS or GFDL results. Both models show very large forcings over East Asia, for example, yet have minimal response there. This is especially clear when comparing the seasonal radiative forcings and climate response (Figure 3.8). Though some of the spatial mismatches could result from a lag in the climate response to seasonally varying forcings, the divergence between the patterns of forcing and response is large even for areas with minimal seasonality in the forcing (*e.g.* Africa, subtropical Asia).



**Figure 3.8** Seasonal average net tropopause instantaneous radiative forcing ( $\text{W m}^{-2}$ ) in 2050 versus 2000 from short-lived species (left column) and surface temperature (K) response (right column) in the GISS and GFDL models. Boreal winter (December-February) is shown in the top two rows, while boreal summer (June-August) is shown in the bottom two rows. The temperature changes at 2050 are 2046-2055 in the GFDL model and 2041-2050 in the GISS model. Values in the upper right corners give the global mean. (Note that the instantaneous forcings shown here for the GFDL model differ from the adjusted forcings show in Table 3.6.).

#### 3.3.4.2 Precipitation, Sea-level Rise, Soil Moisture, etc.

Changes in other climate parameters such as precipitation or sea level due to short-lived species are typically too small to isolate statistically. These would generally be expected to follow the global mean surface temperature change, however, for many of the most important features. For example, the portion of sea-level rise attributable to thermal

expansion of the oceans would be enhanced by ~20-25% due to short-lived species in the GISS and GFDL models. Similarly, the enhancement of precipitation along the equator and drying of the subtropics that is a robust feature of climate models under a warming climate (Held and Soden, 2006) would also be accentuated under the GFDL and GISS models with their significant tropical warming, though probably not under the NCAR scenario. Such a feature can indeed be seen in the GISS response in the Atlantic and Indian Oceans.

On a regional scale, there are some suggestions of trends but statistical significance is marginal for either annual or seasonal changes. The NCAR model shows reductions in winter precipitation due to short-lived species across most of the United States in the 2040s, and reductions in summer precipitation in the southeastern part of the United States. That model also suggests an increase in summer monsoon rainfall over South Asia. In contrast, the GISS model shows slight increases in winter precipitation over the central United States, and a mixed signal in summer (and spring) with increased precipitation over the Southeast and Southwest United States but decreases over the Northeast United States. During fall, precipitation decreases over most of the country. As in the NCAR model, there is an increase in summer (and fall) precipitation over South Asia. In the annual average, the GFDL model shows no statistically significant trend over the United States. Given that significant trends are hard to identify in any of the models, and that the models do not agree on the trends themselves, we believe that it is not possible to reliably estimate precipitation trends owing to short-lived species changes under the A1B storyline.

### 3.3.4.3 Discussion

In the transient climate simulations, three climate models examined the response to projected changes in short-lived species. The results differed substantially among the models. Comparison has shown that the differences in the underlying emissions projections, due to differences between the various Integrated Assessment Models that provided those projections and to assumptions made about emissions not provided by the Integrated Assessment Models, were the dominant source of inter-model differences in projected aerosol trends. These were not the only source of differences, however. For example, the GFDL model's aerosol optical depth is substantially more sensitive to sulfate than the GISS model, with the NCAR model in between. This is partially due to the inclusion of sulfate absorption onto dust being present only in the GISS model. Additionally, the indirect effect of aerosols was included only in the GISS model. Thus, the inclusion of different physical processes played a role in the inter-model differences, and was especially important near 2030, when sulfur dioxide (SO<sub>2</sub>) emissions were near their peak. With the inclusion of the aerosol indirect effect, the GFDL model might yield a substantially larger warming, given that sulfate is the largest contributor to aerosol mass globally and that the GFDL sulfate concentrations decreased beyond 2030.

Differences were also created by the models' differing simulations of the hydrologic cycle, which removes soluble species, and oxidation. Inter-model differences between the GFDL and GISS models in the residence times of aerosols were substantial for sulfate, and differences in the radiative effect of black carbon were also potentially sizeable. In

nearly all cases, however, these were outweighed by emissions differences. This was not the case for sulfate at 2030, nor for tropospheric ozone; differences in the modeled conversion of SO<sub>2</sub> to sulfate and the sensitivity of ozone to NO<sub>x</sub> emissions were larger than differences in projected precursor emissions. Hence, in these cases uncertainties in physical processes, including chemistry, dominate over uncertainties in emissions.

We also reiterate that uncertainties in the aerosol indirect effect and in internal mixing between aerosol types are either not included at all or only partially in these simulations. Sensitivity studies and analysis of the GISS model results indicates that the forcing from reductions in the aerosol indirect effect was roughly 0.1-0.2 W m<sup>-2</sup>, while the inclusion of sulfate absorption onto dust reduced the negative forcing from sulfate at 2030 or 2050 by up to 0.2 W m<sup>-2</sup> (Shindell *et al.*, 2007). These sensitivities suggest that uncertainties in these processes could alter the global mean projected temperature trends by up to 0.1 K at 2030 or 2050, a value comparable to the total temperature trend in that model. Hence without the aerosol indirect effect, the GISS model would likely have shown minimal warming, while without sulfate absorption onto dust surface it would like have shown a substantially greater warming trend (at least at 2030).

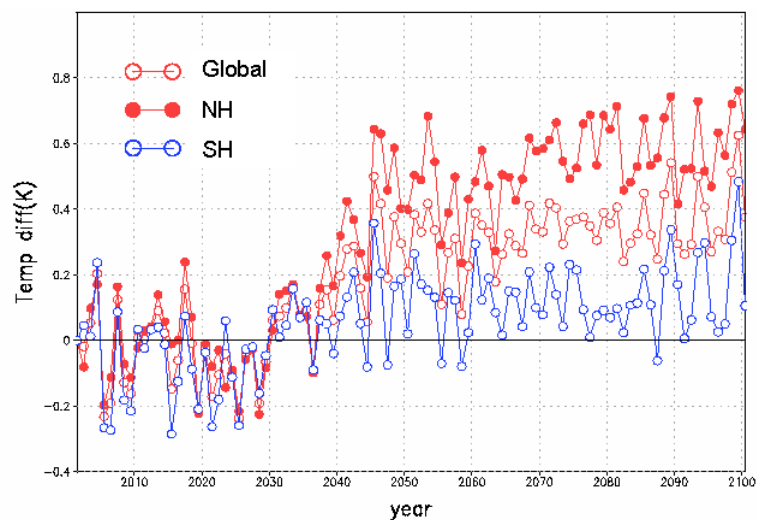
The responses of methane (and other hydrocarbon) emissions and of stratosphere-troposphere exchange to climate change can also potentially have significant impacts on radiative forcing, and these processes were not included in these simulations. As discussed in sections 3.3.3.3 and 3.3.3.4, the resulting changes to radiative forcing could again substantially alter the projected temperature trends. Additionally, given the large

influence of uncertainties in emissions projections, we stress that the magnitude and even the sign of the effects of short-lived species on climate might be different were alternative emissions projections used in these same models. Thus, the response of short-lived species and methane has been only partially characterized by the present study, and substantial work remains to reduce uncertainties and further clarify the potential role of short-lived species in future climate change.

The results clearly indicate that the spatial distribution of radiative forcing is generally less important than the spatial distribution of climate sensitivity in predicting climate impact. Thus, both short-lived and long-lived species appear to cause enhanced climate responses in the same regions of high sensitivity rather than short-lived species having an enhanced effect primarily in or near polluted areas. This result is supported by analysis of the response to larger radiative perturbations in these models for the future (Levy *et al.*, 2007) and the past (Shindell *et al.*, 2007). It is also consistent with earlier modeling studies examining the response to different inhomogeneous forcings than those investigated here (Mitchell *et al.*, 1995; Boer and Yu, 2003; Berntsen *et al.*, 2005; Hansen *et al.*, 2005). This suggests that the mismatch between model simulations of the regional patterns of 20<sup>th</sup> century climate trends and observations is likely not attributable to unrealistic spatially inhomogeneous forcings imposed in those models. Instead, the models may exhibit regional climate sensitivities that do not match the real world, and/or some of the observed regional changes may have been unforced.

### 3.3.5 Climate Simulations Extended to 2100

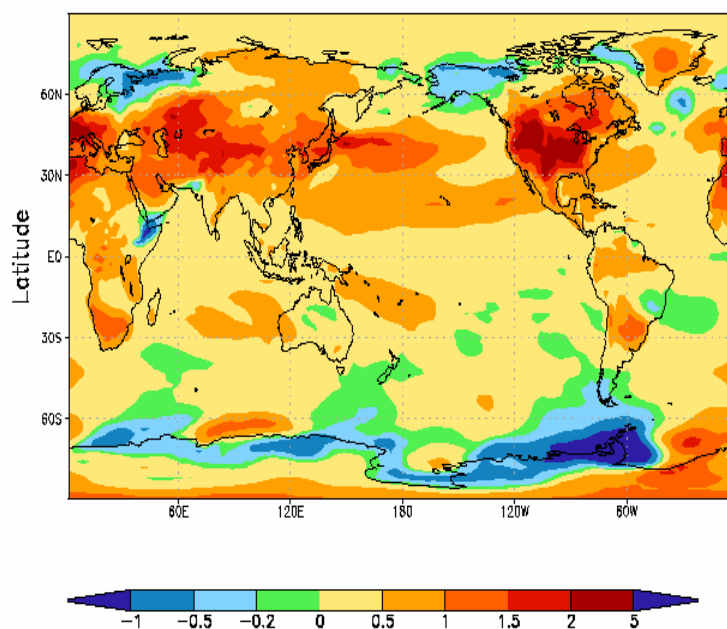
Following the A1B “marker” scenario into the second half of the 21st century for both three-member ensembles, the GFDL simulations (Levy *et al.*, 2007) find significant climate impacts due to emissions of sulfur dioxide (SO<sub>2</sub>), the precursor of sulfate aerosol, (which decrease to ~35% of 2000 levels by year 2100) and of black carbon (scaled to carbon monoxide emissions in the GFDL model) which continue to increase. This is confirmed by their respective radiative forcing values for 2100 in Table 3.6. By 2080 – 2100, these projected changes in emission levels of short-lived species contribute a significant portion of the total predicted surface temperature warming for the full A1B scenario; 0.2°C in the Southern Hemisphere, 0.4°C globally, and 0.6°C in the Northern Hemisphere as shown in the time series of yearly average surface temperature change in Figure 3.9.



**Figure 3.9** Surface temperature change (2000 to 2100) due to short-lived species in the GFDL model.

In Figure 3.10 we examine Northern Hemisphere summer surface temperature change between the 2090s (average over 2091-2100) and the 2000s (average over 2001-2010) due to the changes in emissions of short-lived species between the first decade and the

last decade of the 21st Century. Note the large warming in the Northern Hemisphere mid latitudes with the major hot spots over the continental United States, Southern Europe and the Mediterranean. Eastern Asia, the region with the strongest radiative forcing due to changes in emissions and loading of short-lived species, is not one of the hot spots. The mid latitude warm belt is statistically significant to the 95<sup>th</sup> percentile and the hottest spots are significant to the 99<sup>th</sup> percentile. By contrast, the annual-average and seasonal patterns of change in precipitation due to changes in emissions of short-lived species between the first and last decades of the 21st Century (not shown) are, in general, not statistically significant.

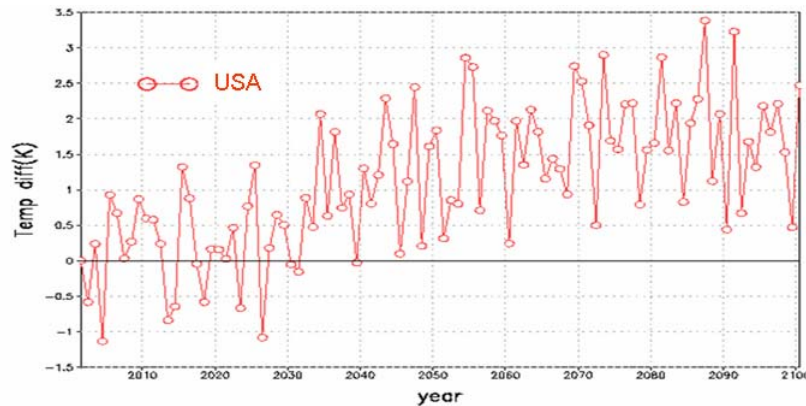


**Figure 3.10** Surface temperature change due to short-lived species during Northern Hemisphere summer for 2100-2091 vs. 2010-2001 in the GFDL model.

We now focus on the large summertime warming over the United States and consider the 21<sup>st</sup> Century time series shown in Figure 3.11. By 2100, the change in short-lived species, primarily the decrease in sulfate and increase in black carbon over Asia, contribute



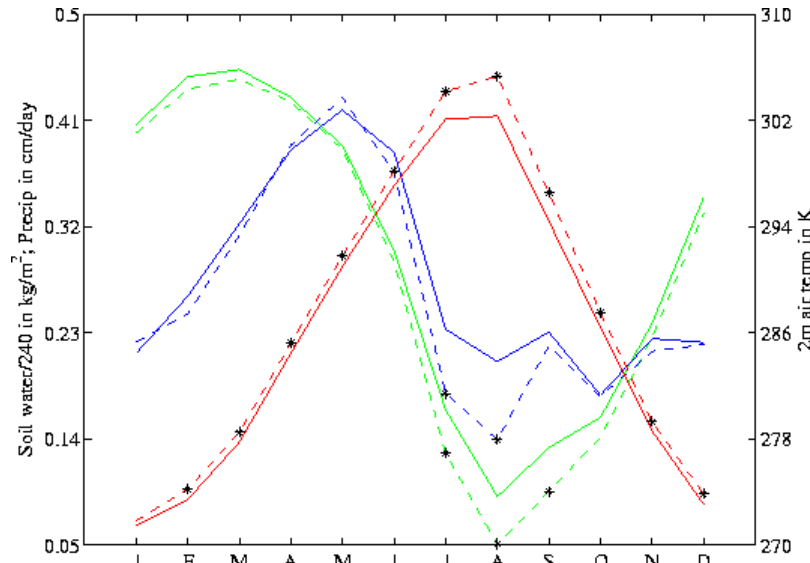
~1.5°C of the ~4°C temperature warming predicted for the continental United States with the effects of changes in both short-lived and long-lived species included.



**Figure 3.11** Surface temperature change (K) due to short-lived species over the continental United States during Northern Hemisphere summer in the GFDL model.

In Figure 3.12, we focus more narrowly on the central United States where the strong summertime warming was predicted for 2100. Monthly-mean area-averaged values of temperature, precipitation, and available root-zone soil water are shown for both the full A1B emission scenario (dashed lines) and the emission scenario with short-lived species levels fixed at 2001 values (solid lines). The values are ensemble averages over the last 40 years (2061-2100) for each simulation. Here the climate model does predict a statistically significant (at the 95% confidence level) decrease in precipitation due to the change in short-lived species (blue curves in Figure 3.12). We next consider root-zone soil water, a quantity that integrates and responds to both temperature and precipitation. There is a statistically significant (at the 95% confidence level) decrease of up to 50% in available root-zone soil water in the Central United States during late summer (July through September), which could have important consequences for United States grain production, and merits future attention. This is the result of a global increase in short-lived species forcing, located primarily over Asia, which in turn results from the large

changes projected by the A1B “marker” scenario for Asian emissions of SO<sub>2</sub> and black carbon.



**Figure 3.12** Monthly-mean time-series of available root-zone soil water (green lines, scaled by a factor of 1/240 for plotting purposes), precipitation (blue lines), and 2-meter air temperature (red lines), averaged over the Central United States (105 to 82.5°W longitude; 32.5 to 45°N latitude). Dashed lines are for the ensemble mean of the A1B experiments, averaged over the years 2061-2100; solid lines are for the ensemble mean of the A1B\* experiments, also averaged over the years 2061-2100. The asterisks represent those A1B monthly average values that are different from their companion A1B\* values at the 95% confidence level.

We also find, as already discussed for year 2050 in Section 3.3.4, that the regional patterns of climate change in 2100 due to changes in emissions of short-lived species are the result of regional patterns in the climate system’s response, rather than regional patterns in radiative forcing. The global patterns of surface temperature change in 2100 are similar for the short-lived species and the well-mixed greenhouse gases with the strongest surface temperature warming occurring over the summer continental US and Mediterranean and the winter Arctic, while the major change in radiative forcing is over Asia (Levy *et al.*, 2007). The predicted summertime warming over the US is greatly enhanced by projected reductions in SO<sub>2</sub> emissions and increased black carbon emissions

and the resulting positive radiative forcings over Asia. In the A1B scenario, this is assumed to be the result of Asian decisions addressing their local and regional air quality. The integrated assessment model projections for A1B assume that SO<sub>2</sub> emissions will be reduced in the future in order to improve air quality, but did not explicitly project carbonaceous aerosol emissions. Scaling future carbonaceous emissions according to carbon monoxide emissions projections does not lead to similar reductions in emissions of these particulates, so that there is an issue of consistency in projecting the influence of future air quality decisions that deserves further study.

### **3.4 Regional Emission Sector Perturbations and Regional Models**

#### **3.4.1 Introduction to Regional Emission Sector Studies**

An additional set of simulations used global models to examine the impact of individual emission sectors in specific regions on short-lived species. This study, in which the GISS and NCAR groups participated, was designed to examine the climate effects of short-lived species in a more policy-relevant way by focusing on the economic activities that could potentially be subject to regulation or reduction in usage (*e.g.* by improved efficiency). We look at reductions in total emissions from a given sector in particular regions (North America and Asia), and do not consider any changes in technology or the relative contributions within a sector. As such, these are more useful for assessing the potential impacts of reductions in total power/fuel usage rather than changes in the mix of power generation/transportation types or in emissions control technologies targeted at specific pollutants.

### 3.4.2 Global Models

The GISS model setup for the regional emission sector perturbation experiments was the same as that used in the transient climate studies (section 3.2 and 3.3; see Appendix 3.1 and 3.2, sections on Goddard Institute for Space Studies). The NCAR regional/sector perturbation simulations used the CAM-chem model (Lamarque *et al.*, 2005b), in which an updated version of the MOZART chemical transport model (Horowitz *et al.*, 2003) is embedded within the Community Atmosphere Model (CAM3, Collins *et al.*, 2006). CAM-chem has a representation of tropospheric chemistry with non-methane hydrocarbons (NMHCs) treated up to isoprene, toluene and monoterpenes. The aerosol simulation in CAM-chem includes the bulk aerosol mass of black carbon (BC, hydrophobic and hydrophilic), primary organic carbon (POA, hydrophobic and hydrophilic), secondary organic carbon (SOA), ammonium and ammonium nitrate, and sulfate aerosols. Further details on the CAM-chem model are found in Appendix 3.2 in the section on National Center for Atmospheric Research.

### 3.4.3 Impact of Emission Sectors on Short-Lived Species

This set of experiments consisted of six simulations each reducing the present-day emissions by 30% in one sector for one region. By using present-day emissions, the results are not tied to any particular scenario. For present-day emissions, the IIASA 2000 inventory, based on the 1995 EDGAR3.2 inventory extrapolated to 2000 using national and sector economic development data, was used (Dentener *et al.*, 2005), as in the GISS simulations described above. The exception to this is biomass burning emissions, which are taken from the Global Fire Emission Database (GFED) averaged over 1997-2002

(Van der Werf *et al.*, 2003) with emission factors from (Andreae and Merlet, 2001) for aerosols. The regions were defined as North America (60-130°W, 25-60°N) and Asia (60-130°E, 0-50°N) and the economic sectors defined according to the IIASA inventory as the domestic sector, the surface transportation sector, and a combined industry and power sector (the NCAR model did not perform the transportation sector simulations).

A control run with no perturbations was also performed to allow comparison. The goal of these simulations was to calculate the radiative forcing from all the short-lived species to identify the relative contribution of the given economic sectors in these two regions. This complements prior work examining the response to a subset of the species included here (*e.g.*, Koch *et al.*, 2007; Unger *et al.*, 2007). As the forcings were expected to be small, we concentrate on simple metrics rather than the climate response. NCAR did not calculate radiative forcing, so we also use aerosol optical depth, which is a good indicator of the radiative forcing from aerosols. All simulations were 11-year runs, with analysis performed over the last 10 years.

The simulations were not performed using a full methane cycle, but the methane response to the imposed perturbations can be estimated by examining the changes in methane's oxidation rate. In these simulations, methane was prescribed at present-day values. Thus any change in methane oxidation is due solely to changes in the abundance of oxidizing agents. The difference in the steady-state abundance of methane that would occur as a result of this oxidation change is a simple calculation ( $[\text{CH}_4]'/[\text{CH}_4] = L/L'$  for the global mean where  $L$  is the methane loss rate and the 'prime' notation indicates the adjusted

amounts). Use of the model's oxidation rate in the perturbation runs fully captures spatial and seasonal variations, and thus provides an accurate estimate of the equilibrium response of methane to the emissions changes. Finally, the radiative forcing resulting from these indirect methane changes is calculated using the standard formulation (Ramaswamy *et al.*, 2001).

We first examine the global mean annual average radiative forcing in the GISS model from the regional perturbations and those by economic sectors (Table 3.10). The effect of the perturbations is generally larger for Asian than in North American emissions. The only radiative forcing from an individual species to exceed  $10 \text{ mW m}^{-2}$  from a North American perturbation is the sulfate forcing from a reduction in industrial/power emissions. In contrast, forcings from sulfate, black carbon, organic carbon and ozone all exceed  $10 \text{ mW m}^{-2}$  in response to perturbations in developing Asia, with the largest response for reductions in black carbon when domestic emissions are reduced ( $-42 \text{ mW m}^{-2}$ ). The spatial pattern of the radiative forcing is also shown (Figure 3.13). The two largest net forcings are in response to changes in North American industrial/power emissions, whose forcing is positive and is dominated by reductions in forcing from sulfate, and the Asia domestic sector, whose forcing is negative and dominated by reductions in black carbon and ozone. The spatial pattern of the aerosol optical depth changes capture the bulk of the radiative forcing in these two cases (Figure 3.14 versus Figure 3.13). The sign is opposite, however, in the case of industrial/power emissions as these dominated by reflective sulfate aerosols, so decreased aerosol optical depth causes positive radiative forcing.

The GISS and NCAR models show very similar patterns of aerosol optical depth changes for these two perturbation experiments. For emissions reduction in the Asia domestic sector, the global mean aerosol optical depth decreases by 0.15 in the GISS model and 0.13 in the NCAR model, while for the North American industrial/power sector the decreases are 0.09 and 0.13, respectively. Hence the aerosol response appears to be fairly robust across these two models. Results suggest that the calculation of radiative forcing from aerosol optical depth introduces an additional inter-model difference that is less than that from the aerosol optical depth calculation (Schulz *et al.*, 2006), so that the total inter-model variation in RF from aerosols is probably on order of 50%. Results for ozone show marked differences, however, with the response of the tropospheric ozone column in the GISS model nearly always a factor of 2-3 greater than in the NCAR model. We believe that these differences primarily reflect the inclusion of the stratosphere in the GISS model, which leads to enhanced forcing as ozone near the tropopause has a particularly large radiative impact. Hence, the ozone radiative forcing is not yet robust to inter-model differences. However, aerosols typically have a larger influence on climate than ozone, so that the net radiative forcing remains a relatively more robust quantity.

We also examine changes in surface pollution levels in these simulations. Changes in surface ozone are typically small, with annual average local reductions of up to about 1-1.5 ppbv in both the NCAR and GISS models in response to reduction in transportation or industrial/power emissions. These increase to levels of 1-3 ppbv during boreal summer. Both these annual and summer increases are statistically significant. Changes in

particulate are larger. In most cases, substantial reduction in surface particulate concentrations result from the regional economic sector emissions reductions. This is especially so in the Asia domestic analysis, where summer sulfate concentrations are lowered by 100-250 pptv locally, and black carbon concentrations drop by 1800-2000 pptv for both summer and annual averages. Smaller air quality improvements are also clear in the response to industrial/power and transportation emissions reductions in both regions. These reductions in particulate are generally quite similar in the two models, with differences of only 5-20% in most cases.

The analysis shows that reductions in surface transportation emissions have a net negative forcing from short-lived species in both regions, primarily due to reductions in black carbon and ozone. As these are both pollutants at the surface, reducing emissions transport offers a way to simultaneously improve human health and mitigate climate warming (though the climate impact is quite small for Asia). The total climate mitigation would of course be larger adding in the effect of reduced emissions of long-lived greenhouse gases. In contrast, industrial/power sector emissions have their largest effect on climate via short-lived species through sulfate, and hence yield a positive forcing. Thus, the net effect of changes in short-lived species to industrial/power emissions reductions will offset a portion of any climate benefit from reduced emissions of long-lived species. The domestic sector presents a more similar picture to that seen for surface transportation. The effects are substantially larger in Asia, however. Hence, reductions in domestic emissions from Asia offer another means to improve human health and mitigate warming. Note that the effects become particularly strong in Northern Hemisphere



summer (Table 3.12), offering a potential path to help mitigating increased summer heat over the Northern Hemisphere continents.

Overall, the Asia domestic emissions offer the strongest leverage on climate via short-lived species. This is partially a result of their magnitude, and partially their occurrence at lower latitudes than North American (or European) emissions. This enhances their impact as photochemistry is faster and incoming radiation is more abundant at lower latitudes. Perturbing the Asia domestic sector in the IIASA 2000 emissions inventory used here yields a much greater effect via black carbon changes than via sulfate changes. This reflects the influence of domestic fuel usage, for which black carbon is the dominant emitted species, and hence reductions from emissions in this sector in particular seem attractive for warming mitigation. As domestic usage and emissions are extremely difficult to quantify in the developing world, further studies of this sector are especially needed to characterize the uncertainty in these emissions. The GISS and NCAR results differ in the magnitude of the aerosol optical depth change resulting from the Asia domestic sector perturbations by only 13%, and this sector/region has the largest influence in both models for both radiative forcing and surface pollution. The stronger aerosol optical depth response in the NCAR model suggests that the RF in that model might be even larger than the  $50 \text{ mW m}^{-2}$  global mean annual average seen in the GISS results.

Further work is required to more thoroughly characterize the robustness of these conclusions across a larger number of models, to explore the impact of aerosol indirect

effects on clouds, and to examine alternative emissions scenarios considering changes in the mix of sources constituting a given sector and the influence of potential technological changes. The latter could be designed to reduce emissions of particular pollutants, while not affecting others. Our results for the radiative forcing from individual species give an idea of the potential impact of such technologies. However, we note that these technologies could also have effects on overall fuel consumption by altering the efficiency of a particular process.

Interestingly, both the transient climate projections and the present-day perturbations find that emissions from Asia are the most important controllers of climate trends or mitigation. Given that the RF reduction from decreases in Asia domestic emissions extends over much of the Northern Hemisphere (Figure 3.13), and the conclusion from the transient climate simulations that the climate response to short-lived species changes is not closely localized near their emissions, it seems plausible that emissions from this region may have as large or larger an effect on other parts of the Northern Hemisphere as changes in local emissions.

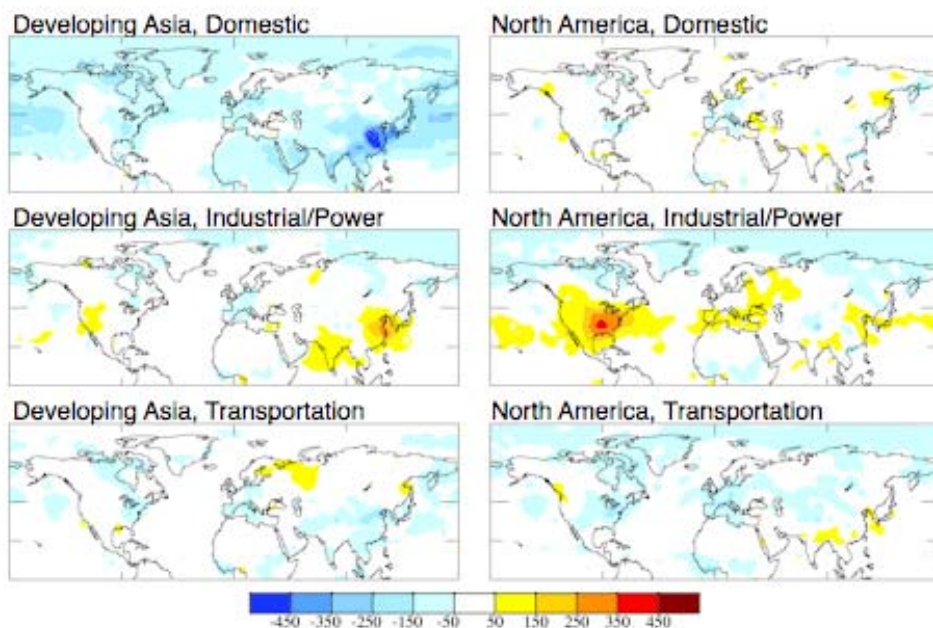
**Table 3.10** Radiative forcing (in  $\text{mW m}^{-2}$ ) from regional emission sector perturbations in the GISS model.

Region	Sector	Sulfate	BC	OC	Nitrate	Ozone	Methane (indirect)	All
North America	domestic	0	-3	2	1	2	1	4
	surface transportation	-3	-5	0	1	-5	4	-9
	industry/power	14	-2	-1	0	5	2	18
Asia	domestic	0	-42	13	1	-12	-2	-41
	surface transportation	2	-8	1	2	-5	7	-2
	industry/power	13	-4	0	-1	-1	5	12

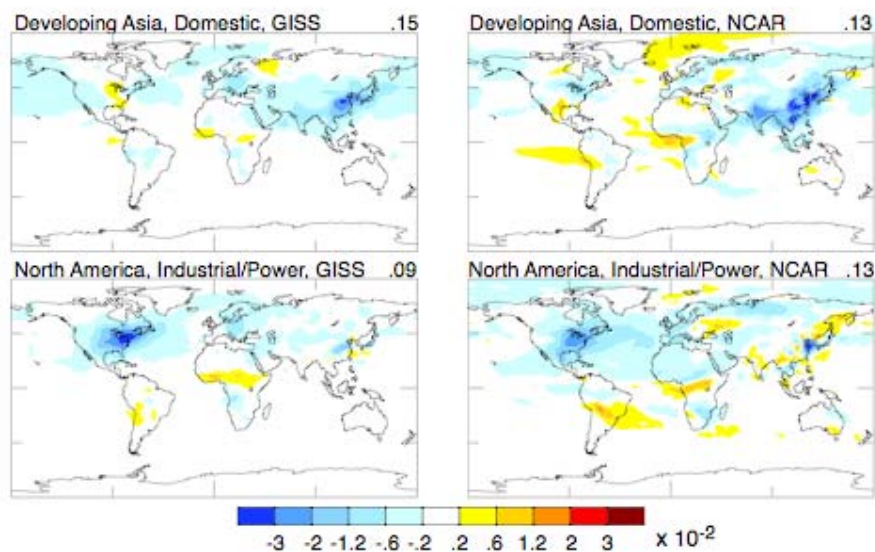
Perturbations are 30% reduction in emissions of all species from the indicated economic sector in the given region. Direct forcings are shown for sulfate, black carbon (BC), organic carbon (OC), nitrate, and ozone. The effect of ozone precursor species on methane is included as methane “indirect”. Note that aerosol indirect effects are not included.

**Table 3.11** Total short-lived species radiative forcing (in  $\text{mW m}^{-2}$ ) as in Table 3.10 but for summer (June-August).

Region	Sector	Total forcing
North America	domestic	6
	surface transportation	-10
	industry/power	34
Asia	domestic	-69
	surface transportation	-3
	industry/power	10



**Figure 3.13** Short-lived species annual average radiative forcing ( $\text{mW m}^{-2}$ ) due to 30% reductions in emissions from the given region and economic sector in the GISS model.



**Figure 3.14** Annual average aerosol optical depth change due to 30% reductions in emissions from the given region and economic sector in the GISS (left column) and NCAR (right column) models. Values in the upper right give the global mean.

#### 3.4.4 Regional Downscaling Climate Simulations

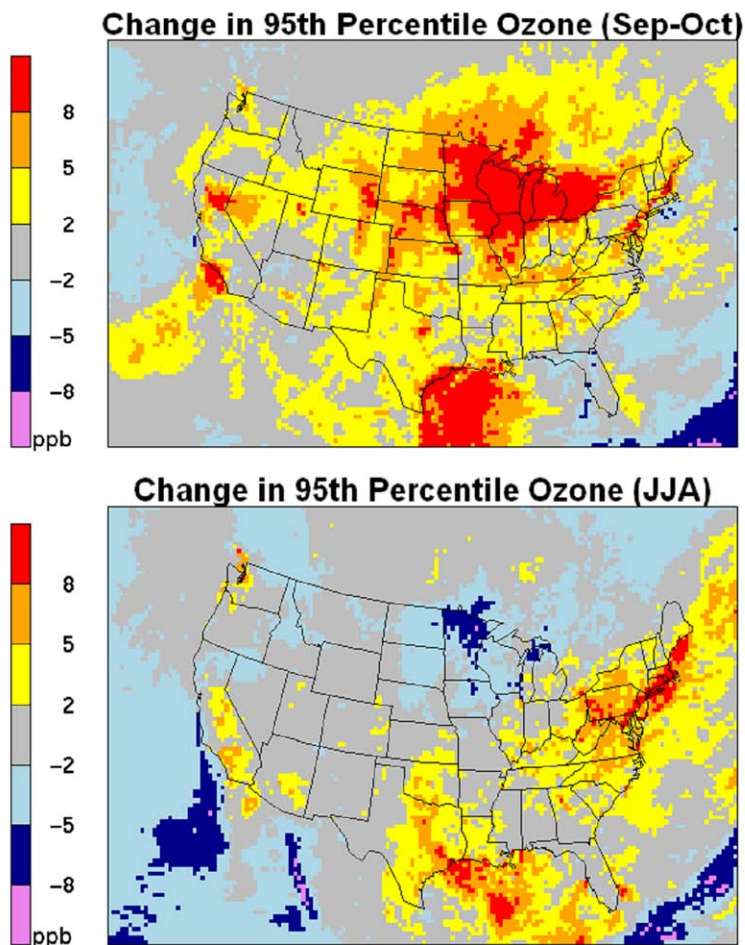
The sector-based simulations presented in Section 3.4.3 suggest that reductions in surface level short-lived species ozone or BC would have a negative RF, while reductions in sulfate aerosols from the utility sector would have a positive RF effect. If concentration levels for these short-lived species (ozone and components of  $PM_{2.5}$ ) exceed threshold standards under the U.S. Clean Air Act, emission control strategies must be developed. Since these short-lived species vary spatially and are affected by local emissions, regional scale models are needed to develop the emission control strategies that meet these standards at county and state levels. To achieve reductions in  $PM_{2.5}$ , emission scenarios often include utility sector emission reductions to lower sulfate aerosol levels. As shown in the previous section, lowering sulfate aerosol concentrations could actually have negative implications for RF and climate temperature increases. Regional downscaling

studies shown in this section suggest that future changes in regional climate could reduce the benefits from anticipated emission reductions on lowering ozone.

Downscaled regional scale climate (RCM) simulations (*e.g.*, Leung and Gustafson, 2005; Liang *et al.*, 2006, Liang *et al.*, 2004) rely on a global climate model to provide boundary conditions for the regional domain as well as the radiative effect of well-mixed GHGs within the domain for the radiation calculations. Regionally downscaled climate simulations are needed by a number of applications that must consider local changes in future climate. Since ozone and PM<sub>2.5</sub> exceedances of regulatory thresholds are substantially affected by local scale changes in emissions and meteorology, several recent studies have used regionally downscaled climate scenarios have been used to study the sensitivity of air quality to potential changes in future climate. The primary purpose of these studies was to study how increases in temperature and other future climate changes could affect ozone and PM<sub>2.5</sub> and potentially decrease the effectiveness of anticipated emission reductions.

Using downscaled RCM simulations from and Gustafson (2005), Nolte *et al.* (2007) have used the EPA/NOAA CMAQ model to test the impact of future (ca. 2050) climate on ozone and aerosols with current emission scenarios. Biogenic emissions, which are meteorologically dependent, were re-calculated under the future climate scenario. Results suggest that future climate changes could increase maximum ozone levels by approximately 10% in some regions (see Figure 3.15). With anticipated emission reductions under U.S. Clean Air Act requirements, these results suggest that future

climate could dampen the effectiveness of these emission controls. Evaluation of ensemble RCM results are essential for this application before quantitative conclusions can be made about the impact of future climate on specific emission control strategies; however, these results suggest that climate change is a factor that needs to be considered in air quality management.



**Figure 3.15** From Nolte *et al.* (2007), the change in ozone at the upper end of the ozone distribution (average of the  $\geq 95^{\text{th}}$  values for each grid) for (2050 - Present) years of simulation under A1B RCM simulations.

These regional downscaling climate studies discussed rely on climate forcing linkages from global climate simulations with future trends for long-lived species including CO<sub>2</sub>,

CH<sub>4</sub>, N<sub>2</sub>O, and chlorofluorocarbon. The influence of short-lived species on future climate has not been included in those studies to date; however, more recent developments are underway to include direct and indirect radiative effects in the regional chemistry model. Based on the known positive radiative forcing effect of ozone, the increases in ozone in response to future climate in Figure 3.15 should have a positive RF effect that could dampen the net negative radiative forcing anticipated from future emission reductions for ozone (see Section 3.4.3).

While regional downscaling climate impacts from short-lived species cannot be directly reported on, future emission scenarios for short-lived species were considered by several regional downscaling studies for the purpose of air quality impacts under future climate. The impact of climate only and then an emission change scenario were tested by (Nolte *et al.*, 2007) and (Hogrefe *et al.*, 2004). As presented in Figure 3.15, results looking only at the climate sensitivity without future changes in emissions suggest that future climate changes could increase maximum ozone levels by approximately 8ppb or 10% in some regions of North America. Looking at future emission scenarios for NO<sub>x</sub> and SO<sub>2</sub> reductions demonstrates that the uncertainty in the future emission scenarios introduces a much larger variation in the air quality conclusions depending on the scenario (Hogrefe *et al.*, 2004; Nolte *et al.*, 2007). Similar to the findings here about short-lived species' impact on climate, the range of plausible air quality impacts from future short-lived emission scenarios suggest very different outcomes, and the future scenarios of emissions for short-lived species have a great deal of obvious, inherent uncertainty.

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## Appendix 3.1 Composition Models

### A.3.1.1 Geophysical Fluid Dynamics Laboratory

Composition changes for the short-lived species from 2000 to 2100 in the GFDL experiments were calculated using the global chemical transport model MOZART-2 (Model for Ozone And Related chemical Tracers, version 2.4), which has been described in detail previously (Horowitz *et al.*, 2003; Horowitz, 2006; and references therein. This model was used to generate distributions of ozone, sulfate, black and organic carbon, and dust for the emission scenarios discussed in Section 3.2.1. Emissions and initial conditions for methane were scaled each decade to match the global average methane abundances specified in the A1B “marker” scenario. The model includes 63 gas-phase species, 11 aerosol and precursor species to simulate sulfate, nitrate, ammonium, and black and organic carbon and five size bins for mineral dust (diameter size bins of 0.2-2.0  $\mu\text{m}$ , 2.0-3.6  $\mu\text{m}$ , 3.6-6.0  $\mu\text{m}$ , 6.0-12.0  $\mu\text{m}$ , and 12.0-20.0  $\mu\text{m}$ ). Hydrophobic black and organic carbon are chemically transformed into hydrophilic forms with a lifetime of 1.63 days (Tie *et al.*, 2005). Different aerosol types are assumed to be externally mixed and do not interact with one another. Sulfur oxidation in the gas phase and within clouds is fully interactive with the gas-phase oxidant chemistry.

The transport in MOZART-2 is driven with meteorological inputs provided every three hours by the middle-atmosphere version of the NCAR Community Climate Model (Kiehl *et al.*, 1998). The meteorology was the same for each decade, thus excluding any feedbacks from climate change on natural emissions and rates of chemical reactions and removal. Thus, natural emissions, such as those of isoprene, dust, and  $\text{NO}_x$  from

lightning, are held constant at present-day levels. Convective mass fluxes are re-diagnosed from the large-scale meteorology using the Hack (1994) and Zhang and McFarlane (1995) schemes. Vertical diffusion within the boundary layer is diagnosed using the scheme of Holtslag and Boville (1993). Tracer advection is performed using a flux-form semi-Lagrangian scheme (Lin and Rood, 1996).

The horizontal resolution is  $2.8^\circ$  latitude x  $2.8^\circ$  longitude, with 34 hybrid sigma-pressure levels extending up to 4 hPa. Photolysis frequencies for clear-sky are interpolated from a pre-calculated lookup table, based on a standard radiative transfer calculation (TUV version 3.0; (Madronich and Flocke, 1998). The values are modified to account for cloudiness (Brasseur *et al.*, 1998), but do not account for effects of the simulated aerosols. Heterogeneous hydrolysis of  $\text{N}_2\text{O}_5$  and  $\text{NO}_3$  on aerosol surfaces occurs at a rate based on the simulated sulfate surface area, with a reaction probability = 0.04 (Tie *et al.*, 2005). Stratospheric concentrations of ozone and several other long-lived gases are relaxed to present-day climatological values.

Dry deposition velocities for gas-phase species are calculated off-line using a resistance-in-series scheme (Wesely, 1989). Deposition velocities for aerosol species are prescribed as by Tie *et al.* (2005). Wet removal of soluble species in and below clouds is included as a first-order loss process, based on the large-scale and convective precipitation rates, as described by Horowitz *et al.* (2003). In-cloud scavenging is based on the parameterization of Giorgi and Chameides (1985), while below-cloud washout of highly soluble species follows Brasseur *et al.* (1998). For gas-phase species, the removal rate

depends strongly on the temperature-dependent effective Henry's law constant. Wet deposition of soluble aerosols (sulfate, hydrophilic BC, hydrophilic OC, ammonium, and nitrate) is calculated by scaling the removal rate to that of highly-soluble  $\text{HNO}_3$ , assuming the aerosols have a first-order loss rate constant equal to 20% of that of  $\text{HNO}_3$  (Tie *et al.*, 2005). This scaling introduces a large uncertainty into the calculation of aerosol burdens. The sensitivity of model results to this scale factor is discussed below (Section 5). Wet removal of dust is calculated using the formulation of Zender *et al.* (2003), with below-cloud scavenging efficiencies of  $0.02 \text{ m}^2 \text{ kg}^{-1}$  for convective and  $0.04 \text{ m}^2 \text{ kg}^{-1}$  for stratiform precipitation.

The ozone and aerosol distributions from these simulations have been evaluated by Horowitz (2006) and Ginoux *et al.* (2006), respectively. Simulated ozone concentrations agree well with present-day observations and recent trends (Horowitz, 2006). Overall, the predicted concentrations of aerosol are within a factor 2 of the observed values and have a tendency to be overestimated (Ginoux *et al.*, 2006). The annual mean surface sulfate concentrations match observed values within a factor 2 with values ranging from  $0.05 \mu\text{g m}^{-3}$  in the remote marine atmosphere to  $13 \mu\text{g m}^{-3}$  in polluted regions. In general, the simulated concentrations are over-predicted in summer and under-predicted in winter. Sulfate mass column and zonal mean profiles are comparable to previous studies, although the global mean burden is about 15% higher. The annual mean concentration of carbonaceous aerosols is generally overestimated in polluted regions by up to a factor of 2. An exception is West Africa where other models show significant loadings of carbonaceous aerosols associated with biomass burning activities during the dry season



while our results do not show any perturbation arising from such activities. The source of this discrepancy seems to be caused in part by the emission inventory in West Africa. The annual mean dust concentration at the surface agrees with the observations to within a factor 2, except over Antarctica where it is underestimated by a factor of 5.

The three-dimensional monthly mean distributions of ozone, black and organic carbon aerosol, and sulfate aerosol from MOZART-2 were archived from simulations for each decade from 2000 to 2100. The results from these simulations were then interpolated to intermediate years and used in the transient climate simulations. The distribution of dust from a present-day simulation in MOZART-2 was used in all years of the climate simulations.

#### **A.3.1.2 Goddard Institute for Space Studies**

The configuration of the GISS composition model used here has been described in detail in (Shindell *et al.*, 2007). The composition model PUCCINI (Physical Understanding of Composition-Climate INteractions and Impacts) includes ozone and oxidant photochemistry in both the troposphere and stratosphere (Shindell *et al.*, 2006). Photochemistry includes 155 reactions. The model calculates the abundances of 51 chemical species, 26 of which are transported by the model's advection scheme. It uses 'lumped families' for hydrocarbons and PANs. Chemical reactions involving these surrogates are based on the similarity between the molecular bond structures within each family using the reduced chemical mechanism of (Houweling *et al.*, 1998). This mechanism is based on the Carbon Bond Mechanism-4 (CBM-4) (Gery *et al.*, 1989),

modified to better represent the globally important range of conditions. The CBM-4 scheme has been validated extensively against smog chamber experiments and more detailed chemical schemes. This scheme was modified for use in global models by removing aromatic compounds and adding in reactions important in background conditions, including organic nitrate and organic peroxide reactions, and extending the methane oxidation chemistry. The revised scheme was then readjusted based on the more extensive Regional Atmospheric Chemistry Model (RACM) (Stockwell *et al.*, 1997), and the modified scheme includes several surrogate species designed to compensate for biases relative to the RACM mechanism. The modified scheme was shown to agree well with the detailed RACM reference mechanism over a wide range of chemical conditions including relatively pristine environments (Houweling *et al.*, 1998).

Rate coefficients are taken from the NASA JPL 2000 handbook (Sander *et al.*, 2000). Photolysis rates are calculated using the Fast-J2 scheme (Bian and Prather, 2002), except for the photolysis of water and nitric oxide (NO) in the Schumann-Runge bands, which are parameterized according to (Nicolet, 1984; Nicolet and Cieslik, 1980). The aerosols component simulates sulfate, carbonaceous and sea-salt aerosols (Koch *et al.*, 2007; Koch *et al.*, 2006) and nitrate aerosols (Bauer *et al.*, 2006). It includes prognostic simulations of DMS, MSA, SO<sub>2</sub> and sulfate mass distributions. The mineral dust aerosol model transports four different sizes classes of dust particles with radii between 0.1-1, 1-2, 2-4, and 4-8 microns (Miller *et al.*, 2006). Most importantly, these components interact with

one another, with linkages including oxidants affecting sulfate, gas-phase nitrogen species affecting nitrate, sulfate affecting nitrogen heterogeneous chemistry via reaction of  $\text{N}_2\text{O}_5$  to  $\text{HNO}_3$ , and sulfate and nitrate being absorbed onto mineral dust surfaces (*i.e.*, the aerosols are internally mixed as coatings form on dust surfaces (Bauer *et al.*, 2006). The latter is described by a pseudo first-order rate coefficient which gives the net irreversible removal rate of gas-phase species to an aerosol surface. We use the uptake coefficient of 0.1 recommended from laboratory measurements (Hanisch and Crowley, 2001), though this value is fairly uncertain.

Phase transformation and removal of soluble species is calculated using a wet deposition scheme in which soluble gases can be removed into either moist convective plumes or large-scale clouds as derived from the GCM's internal cloud scheme (Del Genio and Yao, 1993). During convection, all chemical species are transported along with the convective plumes, with scavenging of soluble species within and below cloud updrafts. In large-scale stratiform clouds, soluble gases are removed based on the fraction of the grid box over which precipitation is taking place. Washout of soluble species is calculated below precipitating clouds. In the case of either evaporation of precipitation before reaching the ground, or detrainment or evaporation from a convective updraft, the dissolved species are returned to the air. Wet chemistry calculations take place in each grid box at each time step, including the coupling with the convection scheme's entraining and nonentraining plumes (which are based on the convective instability in the particular grid box at that time), so are entirely consistent with the contemporaneous model physics. The solubility of each gas is determined by an effective Henry's Law

coefficient, assuming a pH of 4.5. Surface dry deposition is calculated using a resistance-in-series model (Wesely and Hicks, 1977) coupled to a global, seasonally varying vegetation data set as given by (Chin *et al.*, 1996).

The 2000 simulation uses the 2000 emission inventory of the International Institute for Applied Systems Analysis (IIASA), except for biomass burning which is taken from the Global Fire Emission Database (GFED) averaged over 1997-2002 (Van der Werf *et al.*, 2003) with emission factors from (Andrae and Merlet, 2001) for aerosols. The IIASA inventory is based on the 1995 EDGAR3.2 inventory (Olivier and Berdowski, 2001), extrapolated to 2000 using national and sector economic development data (Dentener *et al.*, 2005). Lightning NO<sub>x</sub> emissions are calculated internally in the GCM (5.6 Tg/yr for present-day), and other natural sources are prescribed according to conventional estimates. Dust emissions are constant at 1580 Tg/yr, while isoprene emissions are 356 Tg/yr. Emissions of DMS are 41 Tg/yr.

The simulations described here were run with this composition model included within a 23-layer (up to 0.01 hPa), 4° x 5° horizontal resolution version of the ModelE climate model (Schmidt *et al.*, 2006). This composition model was used for both the transient climate and regional/sector emissions perturbation simulations.

Present-day composition results in the model are generally similar to those in the underlying chemistry and aerosol models documented previously. The model used here does not include the enhanced convective scavenging of insoluble species prescribed in

(Koch *et al.*, 2007). Therefore our carbonaceous aerosol burden, especially in the free troposphere, is nearly double that of (Koch *et al.*, 2007). Comparison with the limited available observations is comparable between the two simulations (a positive bias replaces a negative bias).

### A.3.1.3 National Center for Atmospheric Research

Various methods were used at NCAR to estimate future composition. Present-day tropospheric ozone was taken from calculations performed by Lamarque *et al.* (2005) using the MOZART-2 model; beyond 2000, tropospheric ozone was calculated by T. Wigley using the MAGICC model (<http://www.cru.uea.ac.uk/~mikeh/software/magicc.htm>) forced by the time-varying emissions of NO<sub>x</sub>, methane and VOCs and these average global values were used to scale the present-day distribution. Future carbonaceous aerosols were scaled from their present-day distribution (Collins *et al.*, 2001) by a globally uniform factor whose time evolution follows the global evolution of SO<sub>2</sub> emissions. Future levels of sulfate aerosols were calculated using the MOZART model. Stratospheric ozone changes are prescribed following the study by (Kiehl *et al.*, 1999).

The Model for Ozone and Related chemical Tracers version 2 (MOZART-2) is described by Horowitz *et al.* (2003) and references therein. The model provides the distribution of 80 chemical constituents (including nonmethane hydrocarbons) between the surface and the stratosphere. The model was run at a uniform horizontal resolution of ~2.8° in both latitude and longitude. The vertical discretization of the

3448 meteorological data (described below) and hence of the model consists of 18 hybrid  
3449 levels from the ground to ~4 hPa. The evolution of species is calculated with a time  
3450 step of 20 min.

3451 The tropospheric photolysis rates use a vertical distribution of ozone based on the  
3452 simulated ozone in the troposphere and on the climatology from Kiehl *et al.* (1999)  
3453 above. For each simulation, this latter distribution is updated to reflect the changes in the  
3454 lower stratosphere during the 20th century, affecting only the photolysis rates and not the  
3455 amount of ozone transported from the stratosphere.

3456

3457 The NCAR regional/sector perturbation simulations (Section 3.4) used a version of  
3458 MOZART chemical transport model (Horowitz *et al.*, 2003) embedded within the  
3459 Community Atmosphere Model (CAM3, Collins *et al.*, 2006). This model, known as  
3460 CAM-chem, includes an extension of the chemical mechanism presented by Horowitz *et*  
3461 *al.* (2003) to include an updated terpene oxidation scheme and a better treatment of  
3462 anthropogenic non-methane hydrocarbons (NMHCs). The MOZART aerosols have been  
3463 extended by Tie *et al.* (2001, 2005) to include a representation of ammonium nitrate that  
3464 is dependent on the amount of sulfate and ammonia present in the air mass following the  
3465 parameterization of gas/aerosol partitioning by Metzger *et al.* (2002). In brief, CAM-  
3466 chem simulates the evolution of the bulk aerosol mass of black carbon (BC, hydrophobic  
3467 and hydrophilic), primary organic (POA, hydrophobic and hydrophilic), second organic  
3468 (SOA, linked to the gas-phase chemistry through the oxidation of atmospheric NMHCs  
3469 as in (Chung and Seinfeld, 2002), ammonium and ammonium nitrate (from NH<sub>3</sub>  
3470 emissions), and sulfate aerosols (from SO<sub>2</sub> and DMS emissions). It also considers the

uptake of  $\text{N}_2\text{O}_5$ ,  $\text{HO}_2$ ,  $\text{NO}_2$ , and  $\text{NO}_3$  on aerosols. Results from the CAM-chem model are discussed by Lamarque *et al.* (2005b). A description of sea-salt, updated from Tie *et al.* (2005), is also included. Finally, a monthly-varying climatology of dust is used only for radiative calculations. The CAM-chem model considers only the direct effect of aerosols and the atmospheric model is coupled with the chemistry solely through the radiative fluxes, taking into account all radiatively active gases and aerosols. The horizontal resolution is  $2^\circ$  latitude x  $2.5^\circ$  longitude, with 26 levels ranging from the surface to  $\sim 4$  hPa.

## Appendix 3.2 Climate Models

### A.3.2.1 Geophysical Fluid Dynamics Laboratory

Climate simulations at GFDL used the coupled climate model recently developed at NOAA's Geophysical Fluid Dynamics Laboratory, which has been previously described in detail (Delworth *et al.*, 2006). We will summarize here. The model simulates atmospheric and oceanic climate and variability from the diurnal time-scale through multi-century climate change without employing flux adjustment. The control simulation has a stable, realistic climate when integrated over multiple centuries and a realistic ENSO (Wittenberg *et al.*, 2006). Its equilibrium climate response to a doubling of CO<sub>2</sub> is 3.4C<sup>1</sup> (Stouffer *et al.*, 2006). There are no indirect aerosol effects included in any of the simulations. The resolution of the land and atmospheric components is 2.5° longitude x 2° latitude and the atmospheric model has 24 vertical levels. The ocean resolution is 1° latitude x 1° longitude, with meridional resolution equatorward of 30° becoming progressively finer, such that the meridional resolution is 1/3° at the Equator. There are 50 vertical levels in the ocean, with 22 evenly spaced levels within the top 220 m. The ocean component has poles over North America and Eurasia to avoid polar filtering.

Using a five member ensemble simulation of the historical climate (1861-2003) including the evolution of natural and anthropogenic forcing agents, the GFDL climate model is able to capture the global historical trend in observed surface temperature for the 20th century as well as many continental-scale features (Knutson *et al.*, 2006). However, the model shows some tendency for too much twentieth-century warming in lower latitudes and too little warming in higher latitudes. Differences in Arctic Oscillation behavior



between models and observations contribute substantially to an underprediction of the observed warming over northern Asia. El Niño interactions complicate comparisons of observed and simulated temperature records for the El Chichón and Mt. Pinatubo eruptions during the early 1980s and early 1990s (Knutson *et al.*, 2006). In Figure 7d of Knutson *et al.* (2006), where the model ensemble and observations are compared grid box by grid box, ~ 60% of those grid boxes with sufficient observational data have 20<sup>th</sup> Century surface temperature trends that agree quantitatively with the model ensemble. In general, many observed continental-scale features, including a 20<sup>th</sup> century cooling over the North Atlantic, are captured by the model ensemble, as Figures 7a and 7c in Knutson *et al.* (2006) show. However, the model ensemble does not capture the observed cooling over the southeastern US and it produces a 20th century cooling over the North Pacific that is not observed.

#### **A.3.2.2 Goddard Institute for Space Studies**

The GISS climate simulations were performed using GISS ModelE (Schmidt *et al.*, 2006). We use a 20-layer version of the atmospheric model (up to 0.1 hPa) coupled to a dynamic ocean without flux adjustment, both run at 4 by 5 degree horizontal resolution, as in the GISS-ER IPCC AR4 simulations (Hansen *et al.*, 2007). This model has been extensively evaluated against observations (Schmidt *et al.*, 2006), and has a climate sensitivity in accord with values inferred from paleoclimate data and similar to that of mainstream GCMs; an equilibrium climate sensitivity of 2.6°C for doubled CO<sub>2</sub>.

The modeled radiatively active species influence the climate in the GCM. Ozone and aerosols can affect both the short and long wavelength radiation flux. Water uptake on

aerosol surfaces influences the aerosol effective radius, refractive index and extinction efficiency as a function of wavelength and the local relative humidity (Koch *et al.*, 2007), which in turn affects the GCM's radiation field.

The GISS model also includes a simple parameterization for the aerosol indirect effect (Menon *et al.*, 2002) (see box on aerosol indirect effect). For the present simulations, we use only cloud cover changes (the 2nd indirect effect), with empirical coefficients selected to give roughly  $-1 \text{ W m}^{-2}$  forcing from the preindustrial to the present, a value chosen to match diurnal temperature and satellite polarization measurements, as described in (Hansen *et al.*, 2005). We note, however, that this forcing is roughly twice the value of many other model studies (Penner *et al.*, 2006). The aerosol indirect effect in the model takes place only from the surface through  $\sim 570 \text{ hPa}$ , as we only let aerosols affect liquid-phase stratus clouds.

#### **A.3.2.3 National Center for Atmospheric Research**

The transient climate simulations use the NCAR Community Climate System Model CCSM3 (Collins *et al.*, 2006). This model had been run previously with evolution of short-lived species in the future for the IPCC AR4. The model was run at T85 ( $\sim 1.4^\circ \times 1.4^\circ$  resolution). For this study, a new simulation was performed for 2000-2050 in which ozone and aerosols were kept at their 2000 levels. The equilibrium climate sensitivity of this model to doubled  $\text{CO}_2$  is  $2.7^\circ\text{C}$ .

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